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JAMES G. BECKERLEY, *Editor*
Schlumberger Well Surveying Corporation

MARTIN D. KAMEN, *Associate Editor*
Washington University Medical School

LEONARD I. SCHIFF, *Associate Editor*
Stanford University

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PREFACE

After an interval of three years, the primary cosmic radiation is the subject of a full chapter. Cosmic ray studies, as the chapter in the present volume indicates, now seem to have returned to the basic problem of identifying the origin and sequence of events which take place before terrestial detection, leaving the apparently endless task of finding new particles to those associated with the newer high energy accelerators. Future volumes will review this last aspect of accelerator research. The present volume includes a review of the basic theory of fast nucleon polarization and a review of nuclear excitation by heavy charged particles, both subjects of current activity and importance at the accelerator projects.

Two chapters report on the great volume of nuclear data which have accumulated in recent years. The review on nuclear masses illustrates the substantial labor required to establish numerical data on a firm basis. The review of medium-weight nuclei characteristics is similarly indicative of the immense task in evaluating and analyzing data where only limited theoretical bases exist.

The Geneva Conference on the Peaceful Uses of Atomic Energy took place in August 1955, many months after the present volume was planned. Fortunately (we claim no clairvoyance), the publication at that Conference of many sequestered reports provided two chapters with a surplus of references. As a consequence, nuclear radiation effects in solids and nuclear power reactors are reviewed in a much more complete fashion than anticipated.

The chapter on experimental techniques is concerned with low level counting. Stimuli to increase the sensitivity of nuclear particle detection techniques have come from many quarters, ranging from the archaeologist and his dates to the physicist and his rare nuclear processes. These stimuli have hastened the development of remarkable sensitive methods for measuring very low activities.

Isotope effects are discussed in the review of isotope shifts in atomic spectra. A review of the unusually valuable contributions of the oxygen isotopes to the understanding of chemical processes is also included. Current and increasing interest in radiochemical separations of fissionable material and its products has encouraged us to include a review of the generalized acidity concept.

The radiobiology chapters conclude the volume. As in previous volumes both cellular and vertebrate radiobiology are reviewed.

The substantial assistance of Dr. Robert Hofstadter during the absence of Dr. Schiff is gratefully acknowledged.

J.G.B. M.D.K.
C.D.C. L.I.S.
L.F.C. E.S.
R.E.Z.

ERRATUM

Volume 5

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TIME VARIATIONS OF PRIMARY COSMIC RAYS[1,2]

By V. Sarabhai and N. W. Nerurkar
Physical Research Laboratory, Ahmedabad 9, India

I. INTRODUCTION

The subject of time variations of cosmic rays has grown in importance in recent years with increasing realisation that it will give some clue to the origin of cosmic radiation and of the electromagnetic fields in interplanetary and interstellar space. The close relationship of many of the variations with events occurring on the sun is of great interest in the understanding of solar physics.

A comprehensive review of the status of knowledge concerning cosmic ray intensity variations has been prepared by Elliot (1), but is now about four years old. In the intervening period since the article was written, important experimental as well as theoretical contributions have been made. While individual authors such as Simpson (2), Forbush (3) and Sarabhai *et al.* (4) have reviewed particular aspects of the field principally in the light of work of their own groups, a review of progress in the subject as a whole has not been published recently. It is therefore the purpose of this article to cover principally the period 1952 to the close of 1955, and in treating the subject take over generally from the point where Elliot stopped.

The majority of experimental results dealing with the time variation of cosmic rays relate to studies performed on the surface of the earth after the primary cosmic rays have suffered deflection in the geomagnetic field and interacted with matter in the atmosphere. While experiments made at high altitudes in aeroplanes and balloons reduce the intervening mass of atmosphere, it is nevertheless necessary to make allowance for terrestrial effects before one can interpret the variations in terms of the primary component of the radiation. The highly labourious determination of orbits of charged particles in the earth's magnetic field can now be successfully conducted by the use of modern computing machines, and by elegant analogue experiments using a model terella to represent the magnetised earth. Very significant progress has taken place during the past three years in our understanding of the diffusion of cosmic ray intensity in the atmosphere, thus makking it possible to relate with some confidence the intensity of secondary particles measured under specific conditions with mean energy of the primary spectrum of the radiation. Moreover it is now possible to appreciate the role of meteorological factors on cosmic ray intensity. This is of particular significance for the meson component measured at low levels in the atmosphere and enables corrections to be made for variations of atmospheric origin.

[1] The survey of literature pertaining to this review was completed in March, 1956.

[2] The following abbreviations are used throughout the text: mb. = millibar; m.w.e. = meters water equivalent; L.S.T. = local sidereal time; UM region = unipolar magnetic region

This review therefore deals firstly with these advances which provide a background for the interpretation of studies on the time variations of primary cosmic rays.

The solar and sidereal daily variations of cosmic rays, which arise from solar and galactic anisotropies of the primary radiation as viewed from an apparatus fixed to the spinning earth, have often been referred to in literature without making a distinction with true variations of primary intensity. These daily variations are of interest in connection with the theories of origin of cosmic radiation and of electromagnetic fields in space. Furthermore, the solar anisotropy undergoes changes which are considered with other time variations of the radiation and we therefore devote a section of this review to current knowledge on the anisotropy of primary radiation where new experimental evidence has drastically changed some earlier concepts.

This brings us finally to the subject of time variations observed in primary cosmic rays. Here a mass of new experimental data have become available during the past two or three years. In this review we consider the time variations firstly from the standpoint of the time scale involved. Thus long term changes over periods of one or more years are grouped together. These are followed by an account of short term changes extending from one day to a year. Finally, we describe the solar flare effect which produces changes lasting for some hours. The division is primarily phenomenological in character and does not imply a common physical basis for the effects dealt with in each group.

We next discuss the interpretation of the variations with particular reference to their dependence on the mean energy of primaries, and to their solar and terrestrial relationships. We also consider the important question whether the different types of intensity variations represent increases or decreases or both kinds of changes. At the present moment there are a number of models which have been proposed to explain the established facts concerning cosmic ray time variations. It is impossible within the confines of this review to do adequate justice to all of them. The subjective approach of the authors is responsible for the emphasis on various alternative theories proposed. It is, of course, not claimed that there is unanimity of opinion amongst workers in this field concerning the views expressed in this and a later section of the review dealing with the implications of anisotropy and time variations on theories of the origin of cosmic rays.

2. ADVANCES IN ASSOCIATED FIELDS

2.1 Geomagnetic Effects

While early work on this subject by Lemaitre & Vallarta (5) and Stormer (6) involved labourious numerical calculations of the orbits of charged particles in the dipole field of the earth, the main concern of later investigators was to explain the experimentally observed geomagnetic effects such as the latitude effect and the azimuthal variation of cosmic ray intensity.

Through the use of Liouville's theorem and assuming isotropy of primary radiation it was possible to derive a satisfactory theory which no longer needed the calculation of individual particle orbits. A summary of the results is furnished in an admirable review by Alpher (7). However in the interpretation of the anisotropy of cosmic radiation, and of variations such as the solar flare effect where the isotropy of cosmic radiation is no longer observed, it becomes necessary to revert to a calculation of the orbits of charged particles.

We have to determine the asymptotic coordinates of the velocity vector of a charged particle of given rigidity which arrives in a given direction at a location specified in respect of its geomagnetic coordinates. Schluter (8) and Firor (9) have approached this problem by conducting numerical calculations with electronic computing machines and have confined themselves to primaries of energies extending up to 10 Bev for protons and therefore to observing stations in the middle latitudes. Brunberg & Dattner (10) by an experimental method have conducted a comprehensive investigation covering all primary energies greater than 2 Bev for protons and observing stations situated at different latitudes from equator to the poles. With a well collimated beam of electrons of specified but variable velocity emitted from a miniature gun mounted on a magnetised model of the earth, they have at each latitude determined the asymptotic coordinates of the velocity vector of particles of known rigidity which can arrive at specified angles with respect to zenith in the N-S and E-W planes at the observing station. The values of these coordinates can be read out from graphs published by these authors covering each 10° of latitude. From these coordinates they have prepared diagrams such as the one shown in Figure 1 relating to Kodaikanal on the magnetic equator, wherein the asymptotic coordinates can be read out for various rigidities and directions of arrival on a representation of the celestial sphere. A comparison of some of the results of the terella experiments with those derived from numerical calculations has demonstrated the great accuracy of the former. Brunberg & Dattner's study now forms a most valuable and essential basis for the interpretation of work not dealing with isotropic radiation. For most purposes their results are completely adequate. However, for certain effects it is desirable to have knowledge, which is presently not available relating to directions of arrival in azimuths other than in the E-W and N-S planes.

In the interpretation of high altitude balloon and rocket experiments one has to consider the appreciable contribution of the cosmic ray albedo, and interesting but as yet unpublished work in this direction has been conducted by Vallarta and his co-workers.

2.2 The Diffusion of Cosmic Rays in the Atmosphere

The discovery of new unstable particles and their alternative modes of decay indicates that complicated processes must come into play in the transition of the primary radiation to the secondary radiation observed at various

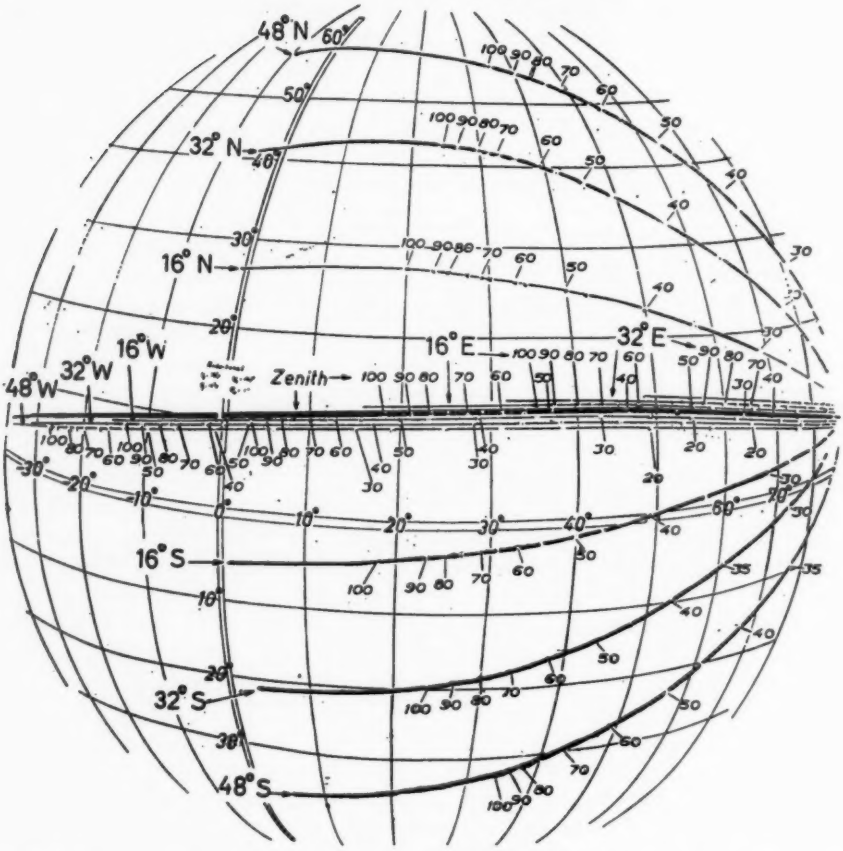

FIG. 1. Diagram showing the asymptotic coordinates of velocity vectors of protons incident at Kodaikanal (geomagnetic latitude 0.3°N). [Brunberg & Dattner (10)].

depths in the atmosphere. Estimates of the energy spectrum of primaries have been made by taking into account the total energy brought in by cosmic radiation at the top of the atmosphere at different latitudes. The primary spectrum has to be related through a specific yield or a generating function to the spectrum of secondaries which are measured at various depths of the atmosphere. Fortunately, for latitude sensitive primaries, it is possible by study of the latitude effect of the measured secondary component at different atmospheric depths, to derive this function without taking specific account of the intervening physical processes.

The latitude effect of the charged particle component at various altitudes in the atmosphere has been studied by the Pasadena workers (11), Pomerantz (12), Berry & Hess (13) and Rao *et al.* (14). Simpson (15) has compared the

latitude effect at 310 g./cm.^{2} depth for the nucleonic component as measured in a neutron monitor and for the meson intensity measured with a counter-telescope. His results are shown in Figure 2. From these data, various workers such as Treiman (16), Fonger (17), Nagashima (18), Dorman (19) and Olbert (20) have calculated the yield of secondaries in a particular energy range at specified depths in the atmosphere produced by primaries of specified energy. The most important conclusions may be summarised in the re-

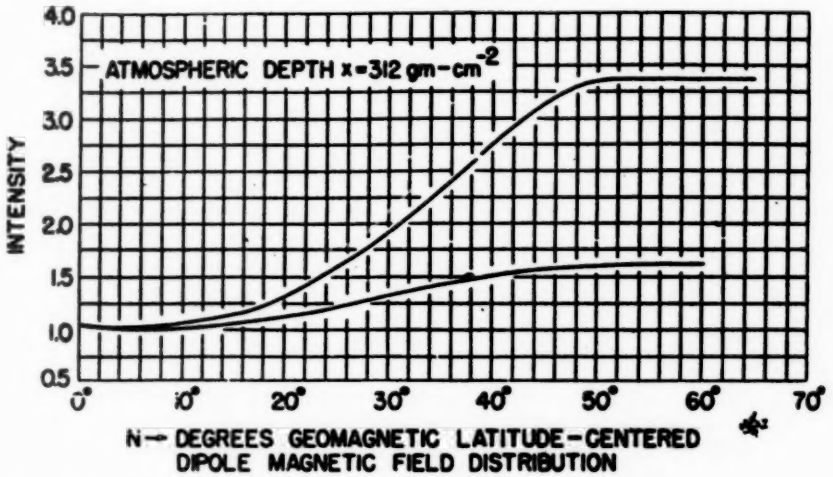

FIG. 2. The upper curve is the neutron latitude effect, the lower curve represents the charged particle component [Simpson (2)].

sults indicated in Figure 3 taken from Fonger. Reasonable extrapolation has been made in order to extend the results to primary energies beyond the latitude sensitive spectrum. The curves enable the intensity measured at 48° latitude in neutron monitors at mountain elevations and in charged particle detectors at sea level to be related to mean primary energy. It would be observed that the nucleonic component as measured by the local production of neutrons at Climax corresponds to a mean primary energy of 7.3 Bev, while the ionization chamber at sea level corresponds to a mean primary energy of 46 Bev. Dorman has made similar calculations and furnished figures from which can be read out the values of coupling coefficients for total ionizing intensity, hard component intensity and neutron intensity at sea level and the total intensity at 4300 meters altitude for the latitudes 0°, 30° and 50° up to primary energies of 1000 Bev.

Since the mean energy of primaries to which a neutron monitor responds is much lower than that for a meson telescope, the dependence of time variations on mean primary energy can be studied by comparing the changes ob-

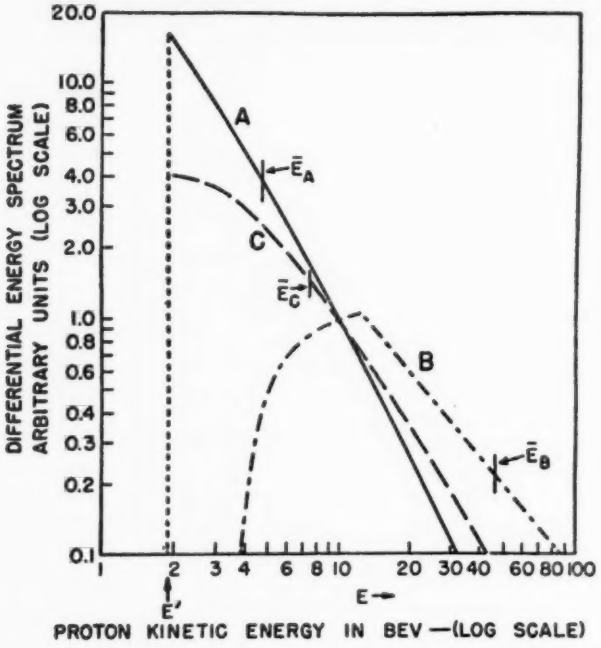

FIG. 3. Time average differential energy spectrum of primary protons arriving from the vertical at geomagnetic latitude $\lambda = 48°$ (Curve A) and the effective primary proton spectra in ionization chamber (Curve B) and neutron detector (Curve C) at the same latitude in the lower atmosphere. All three spectra are normalised to unit amplitude at proton kinetic energy E = 10 Bev. The mean energies of spectra A, B and C are $\bar{E}_A$, $\bar{E}_B$ and $\bar{E}_C$, respectively [Fonger (17)].

served in the two types of instruments. In making this comparison, allowance should be made for difference in the sensitivity of the instruments to radiation in inclined directions to the vertical and for the zenith angle dependence of the observed variation.

2.3 Variations of Atmospheric Origin

A great deal of effort has been devoted to study the effect of meteorological changes on cosmic ray meson intensity. By partial correlation analysis of day to day changes, Duperier was able to derive a regression equation

$$100\frac{\Delta I}{I} = \alpha_P \Delta P + \alpha_H \Delta H + \alpha_T \Delta T$$

connecting the per cent change of meson intensity with the change of barometric pressure ΔP, the change of height of the 100 mb. level ΔH, and the change of temperature of atmosphere between the 100 mb. and 200 mb. levels. Duperier interpreted the barometer coefficient α_p as arising from a mass absorption effect, the decay coefficient α_H as arising from the change

of survival probability of μ mesons when the mean height of meson formation alters, and the positive temperature coefficient α_T as arising from competition between the processes of $\pi-\mu$ decay and the nuclear capture of π mesons which depends on the density of the atmosphere near the level of creation of μ mesons. This equation and the physical meaning ascribed to the coefficients has met with two major difficulties. It was found that the values of the coefficients experimentally determined were not constant. They varied from period to period for the same instrument and they differed according to whether day to day or seasonal changes were considered. Furthermore there was a discrepancy between the lifetime for $\pi-\mu$ decay derived from α_T and what was estimated from other evidence.

Barret *et al.* (21) have pointed out that there is often a relationship bebween ΔH and ΔT. It is then not appropriate to treat these as independent variables. They suggest the adoption of a mean atmospheric temperature which is derived by a process of weighted averaging of the temperatures of successive isobaric levels from ground upwards. They have made observations at a depth of 1574 m.w.e. and since the contribution of $\mu-e$ decay was negligible for mesons corresponding to an energy at production exceeding 10^{11} ev, they were able to study the positive temperature effect arising only due to $\pi-\mu$ decay. They were then able to reconcile the observed positive temperature effect of 0.46 per cent per °C. with the calculated effect using for π mesons a mean lifetime $T=2\times10^{-8}$ seconds in conformity with evidence from other sources. Sherman (22) finds at a depth of 846 m.w.e. an α_T which is smaller than what was expected according to the calculation of Barrett *et al.* for an appropriate energy of μ mesons. Even though this constitutes some evidence for a $\kappa-\mu$ decay process he points out that within the statistical accuracy of the determination the value is not inconsistent with a $\pi-\mu$ decay scheme alone.

The differences in the estimates of α_H and α_T when meteorological changes of a day to day and seasonal character are considered, have been examined by a number of workers. Neglecting $\pi-\mu$ decay, and with a production spectrum of μ mesons as given by Sands (23), Olbert (24) has studied the change of survival probability of μ mesons. He has pointed out that even if the mean height of meson formation remains constant, the survival probability of the mesons depends on the distribution of mass along their path. If there is a proportionately greater mass encountered towards the end of the range, there is correspondingly a reduced ionisation loss and degradation of energy during the earlier part of the path of the meson, thus increasing the survival probablity in the atmosphere. Since there is often an increase of density in the lower layers accompanied by a decrease of temperature in these layers and an increase of temperature at high levels, this effect due to the change of distribution of mass in the atmosphere shows itself as an apparent positive temperature effect of the type observed by Duperier. Olbert has shown that such an effect can be as large as half the value of α_T observed by Duperier. Olbert has further demonstrated that when one considers a regression equation

connecting ΔI with ΔH and ΔP only, neglecting the role of ΔT, the contribution of ΔT gets included in the term containing ΔH. As the relationship between ΔH and ΔT differs in seasonal and day to day changes, he was able to reconcile the apparent differences in the values of α_H when two different types of variations were considered by Elliot & Dolbear (25).

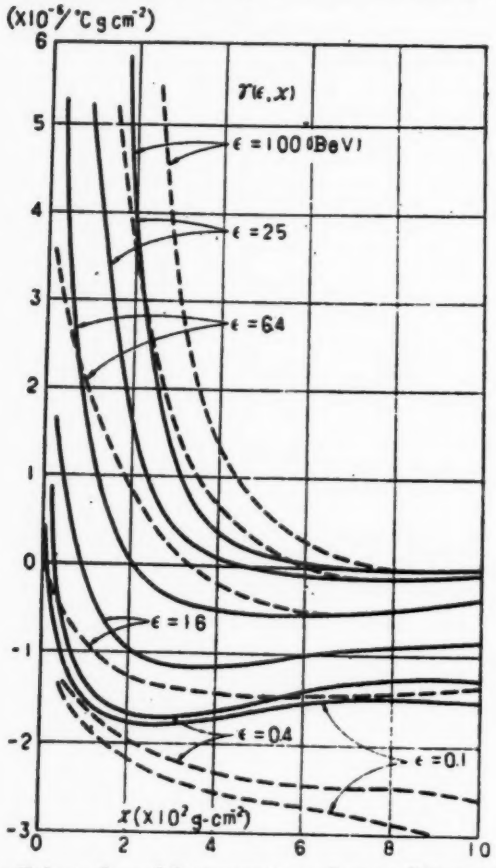

FIG. 4. The coefficient of partial temperature effect at different depths in the atmosphere [Maeda & Wada (26)].

Maeda & Wada (26) have extended Olbert's work by taking into account the process of $\pi-\mu$ decay. They have calculated the contribution to α_T of $\pi-\mu$ and $\mu-e$ decay processes taking into account the temperature variations at successive levels in the atmosphere. They find that the contribution of $\mu-e$ decay is almost constant irrespective of depth from the 50 mb. level downwards. However, the contribution of $\pi-\mu$ decay decreases exponentially with increasing atmospheric depth. They have presented the results graphically in a very convenient form as shown in Figure 4. It should be kept

in mind that the values of α_T that one derives are to be used in conjunction with mass temperature of the atmosphere, and not with the temperature of individual layer as used by Duperier.

Hayakawa *et al.* (27) have extended the work of Maeda & Wada by considering the effect of nucleonic cascades on the production of μ mesons. They find that for the purpose of the $\pi-\mu$ decay process, it is more appropriate to consider the temperature of the 200 mb. level than the average temperature between 50 mb. and 200 mb. They furnish more accurate estimates than Barret *et al.* for the increase of α_T with increasing energy of μ mesons. The calculated values of α_T agree better with observations than the estimates of Maeda & Wada.

Trefall (28) has analysed the implications of various assumptions made in Duperier's regression equation in terms of the interpretation given to the coefficients that are derived from it. He points out that an apparent positive temperature effect can be obtained if the chosen level at which one considers ΔH differs from the true mean level of meson production. If for instance ΔH is taken in relation to the height of the 50 mb. level and the true mean level of production is below it, the distance between the true level of meson production and ground is not only controlled by the height of the assumed level of 50 mb. but by the temperature between it and the 50 mb. level. Thus an apparent positive temperature effect results from the change of survival probability of μ mesons. Thus Trefall was able to explain Duperier's observation that α_T was larger when ΔH was taken at the 50 mb. than at the 100 mb. level.

Trefall (29) has recently pointed out that the survival probability of μ mesons would alter with change of pressure due to ionisation loss. This loss would make a contribution to the barometric coefficient α_p just as Olbert had shown earlier that it would make a contribution to the temperature coefficient α_T. Thus the interpretation of α_p as being due to a pure mass absorption effect requires modification. Trefall estimates that up to a third of this coefficient may arise from the change of survival probability.

It is evident now that the regression equation of the type used by Duperier is inadequate to describe fully the atmospheric effects. In physical terms, his coefficient α_p includes effects due to mass absorption and $\mu-e$ decay, the coefficient α_H relates to $\mu-e$ decay while α_T relates to $\pi-\mu$ decay as well as to $\mu-e$ decay. Wada & Kudo (30) have suggested that by taking the height H to represent the difference between the height of mean level of meson production and an isobaric level near the ground, the mean temperature of the lower atmosphere is taken care of. Moreover this H is independent of sea level pressure. Using this H and a mean atmospheric mass temperature in conjunction with an appropriate temperature coefficient, a satisfactory modification to the original regression equation can be derived. This modified equation has the advantage of being applicable to diverse types of changes of pressure and temperature in the atmosphere, without requiring change of values of coefficients.

The correction of the daily variation of meson intensity for the removal of the effects of atmospheric origin has been considered by several workers. Sarabhai *et al.* (4, 31) have discussed the expected contributions due to daily variation of pressure, of heights of isobaric levels and of temperatures of various levels in the atmosphere. Except at very high altitudes the diurnal heating of the atmosphere is important only up to about 2 Km. above the ground. Near the mean level of meson production there is nonsignificant and small diurnal change of temperature or of the height of isobaric levels. The amplitude of the semidiurnal oscillation of the atmosphere at this level does not exceed a few meters even at the equator. Thus in the daily variation there is expected to be little contribution of the change of height of isobaric levels or of upper air temperature. Maeda (32) has also examined this problem from a number of different aspects. He concludes that there is a small diurnal variation of amplitude about 0.05 per cent with maximum at 1400 hours which he considers to be of atmospheric origin. This is in addition to the contribution of the barometric pressure.

The effect of meteorological factors on the nucleonic component is relatively less complicated than for the meson component where diffusion in the atmosphere involves unstable particles of short life time. Simpson *et al.* (33) have shown that the contribution to the counting rate of a neutron monitor from the local production of neutrons by π and μ mesons is negligible compared to the contribution of nucleons within the nucleonic cascade in the atmosphere. The temperature coefficient due to secondary mesons has been estimated to be less than -0.02 per cent per °C. at the equator and less than -0.006 per cent per °C. at the latitude of 50°. This leaves a mass absorption effect to be corrected for, and Simpson *et al.* have determined the value of 0.96 per cent per mm. of Hg in good agreement with the values reported by Adams & Braddick (34), van Heerdan & Thambyahpillai (35) and Rose & Katzman (36).

2.4 Anisotropy of Primary Cosmic Radiation

2.41 *Solar anisotropy.*—In the past there has been a great deal of misinterpretation of the experimental determinations of the daily variation of meson intensity. This has led to erroneous estimates of the anisotropy of the primary radiation. The daily variation was generally determined with instruments measuring intensity from as wide a range of directions as was possible. This helped to increase the measured intensity and reduce sampling errors. To improve the statistical significance of the determinations, it was customary to take the average of data extending over as large a period as was possible. A small effect with an amplitude of about 0.2 per cent was then observed. However, uncertainties in regard to corrections to be made for the daily variation of meteorological elements led many workers to question the validity of drawing any conclusions regarding the anisotropy of the radiation. Attempts were made by Malmfors (37) and by Elliot & Dolbear (25) to overcome this difficulty by making studies with directional telescopes point

ing to two different directions inclined equally with respect to the vertical. From the difference observed in the daily variations in the North and South directions, it was concluded that there was an anisotropy of primary radiation. On assuming that the North telescope pointing to a fixed direction in space would not be affected by the anisotropy, the difference between north and south telescopes at stations like Manchester and Stockholm was erroneously interpreted as the daily variation corresponding to the anisotropy in the south direction. Sarabhai & Kane (38) have drawn attention to this and suggested that the observed N-S difference can arise from a geomagnetic effect.

As far back as 1950, experimental evidence was not lacking to show that an over-simplification was being made in dismissing evidence connected with the daily variation of meson intensity as being mainly due to effects of terrestrial origin. Ehmert & Sittkus (39) had revealed the difference in amplitude between the daily variations simultaneously measured by an ion chamber of omnidirectional sensitivity and by a vertical counter telescope having a more limited directional sensitivity. Sekido *et al.* (40) had pointed out that the amplitude of the daily variation increased from 0.19 per cent to 0.24 per cent when the semiangle of the measuring telescope in the East-West plane was reduced from 40° to 12°. During the past five years, a series of investigations have been made by workers at Ahmedabad with the object of studying the influence of experimental conditions, such as the characteristics of the apparatus and the latitude and elevation of the observing station, on the nature of the daily variation of meson intensity. Sarabhai *et al.* (4) have shown that at low latitudes the 12 month mean daily variation of meson intensity measured with telescopes with semiangles of 22° in the E-W plane had at different periods of time a diurnal amplitude of 0.3 to 0.8 per cent which is considerably greater than the amplitude observed with the ion chamber at Huancayo. As explained earlier a daily variation of meson intensity of comparable amplitude is hardly expected to be of atmospheric origin. Thus it was concluded that the observed daily variation could in fact be attributed to the anisotropy of cosmic radiation.

Sarabhai *et al.* (41) have conducted experiments where the reduction of the semiangle of the telescopes in the E-W plane may be considered to have been pushed to a practical lower limit. Conscious of the long term changes in the nature of the daily variation, these authors have taken care to ensure that comparisons are made only for data from instruments which have operated for a common period of time. Simultaneous measurements made with instruments with differing semiangles of opening from 15° to 2.5° in the E-W plane reveal a remarkable increase from 0.6 per cent to 1.2 per cent in the amplitude of the daily variation with decrease of semiangle from 15° to 5°. The daily variation measured with each apparatus is shown in Figure 5.

Firor *et al.* (79) have examined the daily variation of the nucleonic component measured with neutron monitors at different latitudes. Averaged

over a large number of days, the daily variation has peak to peak amplitude of about 1.0 per cent which is comparable to the amplitude observed at low latitudes in meson telescopes of moderate semiangle. There is no great latitude dependence of the amplitude of the daily variation. In this respect experience derived from ion chambers of the Carnegie Institution is similar.

Working on the tenative conclusion that the daily variation of meson intensity, after making suitable correction for the barometric effect, can be related to the anisotropy of primary cosmic radiation which is under solar control, Sarabhai & Kane (42) tried to discover whether, as in the many other effects connected with the sun, there is any relationship of the 12

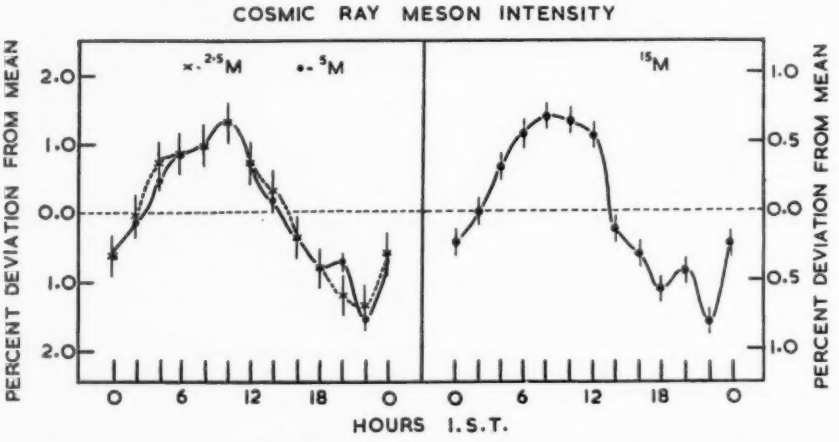

FIG. 5. Comparison of daily variations measured with telescopes of 2.5°, 5° and 15° semiangles in the E-W plane with 19° semiangle in the N-S plane in each case. Indian Standard Time (I.S.T.) is 40 minutes in advance of local time [Sarabhai, Nerurkar & Bhavsar (41)].

month mean anisotropy with the solar cycle of activity. The large changes first observed by them in the 12 month mean daily variation, which are discussed elsewhere in this review, constitute weighty evidence that the association of the daily variation of meson intensity with the anisotropy of primaries is substantially correct. It would indeed be difficult to associate any large contribution of atmospheric origin to the daily variation of meson intensity since meteorological elements are not known to undergo large long term changes of the type observed in cosmic rays.

Concerning the solar anisotropy of the high energy spectrum of the primary radiation, evidence is available from measurements made at great depths underground and with large atmospheric showers. The intensity of the measured radiation being very small, the statistical significance of the determinations is poor in most cases. MacAnuff (43) has reported at a depth of 60 m.w.e. an amplitude of about 0.2 per cent while Sherman (22) at a

depth of 846 m.w.e. and Barrett *et al.* (21) at 1574 m.w.e. exclude the existence of a diurnal variation exceeding 1 per cent in amplitude. Farley & Storey (44) have shown that the daily variation has an amplitude of 1.4 per cent ± 0.25 per cent in investigations on large atmospheric showers. This confirms the absence in the primary spectrum extending upwards from 10^{12} ev of any large solar anisotropy which could produce a daily variation greater than one or two per cent.

The anisotropy of heavy primaries has been studied in balloon experiments by Anderson *et al.* (45), McClure & Pomerantz (46) and Yngve (47). The anisotropy of the very low energy spectrum has similarly been studied by Bergstralh & Schroeder (48) and Dawton & Elliot (49). The experiments are difficult to perform and present even greater difficulties of interpretation. While a daily variation as large as 25 per cent was reported by Yngve, other authors have failed to find a daily variation of amplitude larger than about 1.4 per cent. Since there is great variability of anisotropy from day to day, as is described later, observations made in balloon flights are not strictly comparable amongst themselves and with results reported from ground level stations which relate to the average of data over a large number of days.

2.42 *Galactic anisotropy.*—The study of the sidereal time variation of cosmic rays is related to the search for a galactic anisotropy in the primary radiation. If cosmic rays come from great distances, it would be most appropriate to examine the sidereal time variation of particles which travel almost with the velocity of light and constitute the very high energy region of the primary spectrum. Barrett *et al.* (21) at a depth of 1600 m.w.e. and Sherman (22) at a depth of 846 m.w.e. have estimated that the sidereal daily variation does not have an amplitude exceeding 2.0 per cent and 0.5 per cent respectively. With extensive air showers of primary energy greater than 10^{14} ev, Hodson (50) at Manchester reports an amplitude of 1.15 ± 0.61 per cent and a maximum at 2330 hours L.S.T. Daudin & Daudin (51) for an energy of 6×10^{14} ev indicate an amplitude of 0.39 ± 0.13 per cent at 2200 hours L.S.T. Cranshaw & Galbraith (52) indicate an amplitude of 4.9 ± 1.5 per cent at 1030 hours L.S.T. for an energy of 5×10^{16} ev but an amplitude less than 0.6 per cent for energy up to 10^{16} ev. The most significant result comes from Farley & Storey (44) from Auckland at 37°S. latitude. They get an amplitude of 1.5 ± 0.23 per cent at 1700 hours L.S.T., within half an hour of passage overhead of the galactic plane. The observation of the galactic centre is most favourable in the southern hemisphere and this may account for the discovery of the large sidereal time variation at Auckland. These results taken in conjunction with the observations at Manchester and the Pic du Midi and neglecting those of Cranshaw & Galbraith may be taken to indicate that there is now some evidence for an anisotropy along the plane of the Milky Way, and directed towards the galactic centre. However it is important to realise that in an investigation made by Clark (53) at Boston, no evidence could be found for the existence of any preferred regions or point sources from which there was enhanced intensity of cosmic

radiation within the statistical significance of the observations. It is important to try to confirm the results at Manchester and at the Pic du Midi with more observations at Boston which is also at an intermediate latitude in the Northern hemisphere.

Sekido *et al.* (54) have reported remarkable results in their search for point sources with narrow angle alti-azimuth telescopes inclined to the vertical at 80° to 85°. They give the first reported evidence for a point source of cosmic radiation. The source is in the region of declination 0° and right ascension 5h 30m with an intensity which is 20 per cent of background intensity in the same direction.

3. TIME VARIATIONS

3.1 Long Term Changes of Primary Radiation

3.11 *Changes of total intensity.*—One of the most remarkable variations that has been discovered recently is the change of general intensity of cosmic radiation correlated with the change of solar activity of sunspot numbers. This change of 12 month mean intensity measured by the Carnegie Institution stations was shown by Forbush (3) and by Glokova (55) to be of the order of four per cent between sunspot minimum and sunspot maximum and to be present to almost an equal degree in all four stations ranging in latitude from the equator to 80°N. Figure 6 taken from Forbush demonstrates this

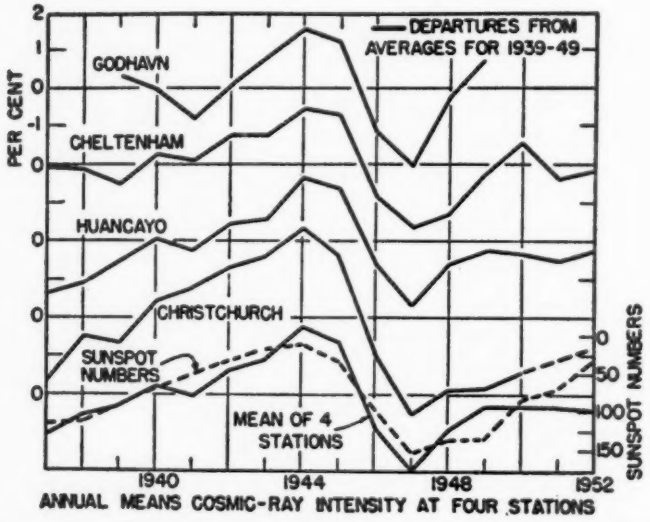

FIG. 6. Annual mean cosmic ray intensity at four stations [Forbush (3)].

change. Forbush has separated his data for magnetically quiet and disturbed days and shows that the change of intensity is present almost equally in both the groups. This is indicative of the fact that the change is not produced

in a simple way by a series of magnetic storm type decreases occurring with varying frequency during different periods of the solar cycle.

Evidence of similar changes at high altitudes is provided by balloon experiments conducted by Neher (56). Corresponding to the change of only four per cent observed at sea level, Neher shows that at the latitude of 85° to 88°N. there was a change in total ionisation of about 50 per cent at an altitude of 70,000 feet from the sunspot maximum year of 1937 to the sunspot minimum year of 1954. Meyer & Simpson (57) in aeroplane flights at 30,000 feet have reported a change of 13 per cent in intensity measured by neutron monitors from 1948 to 1951 compared to a 1 per cent change observed at sea level.

Since high altitude experiments do not provide continuous data of the type that are available at sea level, it is impossible to follow the detailed trend of changes in the low energy component that can only penetrate at high levels of the atmosphere at intermediate latitudes. However, it is worthwhile to draw attention to the fact that flights made by Neher (58) at Bismarck in 1940, when the sun was by no means near the minimum of its activity, reveal an intensity in the upper atmosphere even greater than during the year 1954 of minimum activity. A possible solution of this difficulty has been suggested by Nerurkar by pointing out that while the solar activity during the flights in the years 1940 and 1954 was no doubt higher in 1940 than in 1954, there were periods about three months prior to the flights in each year, for which the solar activity was greater in 1954 than in 1940. Since the high altitude experiments refer to the intensity for a few days and not to the time average of intensity over a period of one year as taken by Forbush, it is conceivable that the anomaly referred to above may only be an apparent one, provided that there is a time lag of about three months between solar activity and changes of intensity. Nerurkar has looked for a time lag of this nature at Huancayo during sunspot minimum when it is expected to be most detectable because day to day fluctuations of cosmic ray intensity are not prominent. He finds that the correlation between monthly mean cosmic ray intensity and monthly mean K_p, indicative of solar activity in the equatorial plane, improves from 0.5 to 0.9 during 1944 and 1945 when the cosmic ray time series is shifted earlier by three months with respect to the K_p time series.[3] The above explanation must be considered tentative until it is confirmed by comparison over longer periods.

3.12 *Change of "knee" of the latitude effect.*—Neher (59) has summarized experimental data extending up to 1950 concerning the measurement of the latitude effect of cosmic rays at various altitudes. A most remarkable feature that emerged from these studies was the absence of a low energy component

[3] K_p is the geomagnetic three-hour-range index introduced by the International Association of Geomagnetism and Aeronomy. It is expressed on a scale of 28 steps and is currently computed from the K-indices of 11 magnetic observatories spread all over the earth. It gives a measure of disturbance in the earth's magnetic field over a three hour interval (IATME Bulletin No. 12e, 1951).

of primary cosmic radiation. The "knee" in the latitude effect indicated that beyond a latitude of about 55° there was no further change in the total cosmic ray intensity even at the highest altitudes that could be reached with balloons. The sharp cut off in the cosmic ray primary spectrum at an energy of about 0.8 Bev for protons corresponding to this "knee" has presented a difficult problem for interpretation.

In attempting to study the finer features of the latitude variation of intensity at high altitudes in high latitudes beyond the "knee," Neher *et al.* (60, 61) profiting by experience gained about the appreciable day to day changes in the intensity of cosmic rays, have since 1951 made simultaneous flights of balloon borne instruments at different latitudes. Thus they have developed a technique to study the "knee" independently of general fluctuations in cosmic rays. Determinations of the "knee" carried out by workers such as Pomerantz (12) and Van Allen & Singer (62) have not involved simultaneous observations at various latitudes and therefore are subject to an appreciable uncertainty as to the conclusions that can be drawn. Figure 7 from Neher shows the "knee" of the intensity versus latitude curves for an atmospheric depths of 15 g./cm.^{2} during 1937, 1951, and 1954. The figure illustrates clearly the shift of the "knee" to higher latitudes from 1937 to 1951. The low energy cut off for protons at 0.8 Bev during 1951 is absent during 1954 when a very large intensity of soft particles down to energies of 0.15 Bev for protons is measured and there is no evidence of a "knee." Meyer & Simpson (57) have reported that from 1948 to 1951, the "knee" of the latitude effect has shifted by 3° from 55 to 58°N. Simpson did not observe much change between 1951 and 1954, but since the results of Neher *et al.* show that this change was almost entirely due to the influence of particles, if protons between 0.15 and 0.8 Bev, it is to be expected that these would produce little effect at 30,000 feet altitude where Simpson's observations were carried out.

The investigation of very low energy particles in the range of 20 Mev to 170 Mev by means of single counters sent aloft in rockets has been done by Meredith *et al.* (63). At the present moment, interpretation of their results is beset with difficulties on account of uncertainty in the contribution of the albedo and of the geomagnetic effects as applied to streams of low energy particles at very high latitudes. Ellis *et al.* (64) point out that during 1953 there was a cut off for heavy primaries at about the same latitude as for protons. This indicates a cut off dependent on the rigidity rather than the energy of the primaries. The results, which require confirmation during other years, therefore favour a mechanism involving magnetic fields as being responsible for the cut off.

3.13 *Changes of solar anisotropy.*—Within the past three years large worldwide changes of the 12 month mean daily variation of meson intensity have been discovered by Sarabhai & Kane (42). By an examination of data from 1937 to 1946 of Forbush & Lange (69) they drew attention to correlated changes of amplitude and of time of maximum of the diurnal component of the daily variation at Christchurch, Cheltenham, and Huancayo. They

pointed out the relationship of these changes with the 11 year cycle of solar activity and with the magnetic character figure, and showed that correlations improved during sunspot minimum. From this they concluded that the changes of daily variation were related mainly to activity in the equatorial belt of the sun. They also suggested that the anisotropy of the radiation was connected with continuous emission of cosmic rays from the sun, but this view does not now appear tenable in the light of further evidence to be discussed later.

Thambyahpillai & Elliot (65) have confined attention to the time of

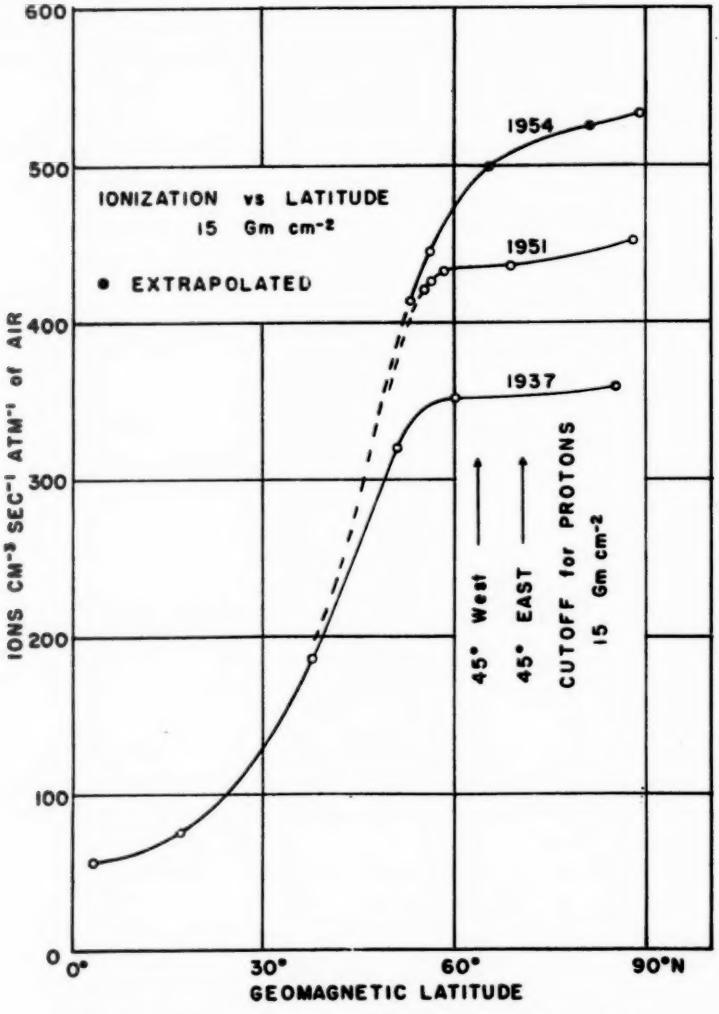

FIG. 7. Long term change of latitude effect at 15 gm./cm.² atmospheric depth. Neher (56).

maximum of the diurnal component of the daily variation. After showing the correlated changes occurring in it at the Carnegie Institution stations, they have combined data from various sources to study the trend of change from 1932 to 1952. The data relate to different types of instruments and in putting them together no allowance has been made for a possible difference in the magnitude of the effect that would be recorded under the differing experimental conditions. However they were led to a general conclusion that the change probably followed a 22 year cycle of solar activity.

Steinmaurer & Gheri (66) have recently compared data taken over groups of years over the past 23 years. They show that during sunspot minimum in 1933, the time of maximum of the diurnal component was at about 7:00 A.M., comparable with the time of maximum about 20 years later in 1953–54, but was earlier than the time of maximum 11 years later in 1944. This indicates again a relationship with the 22 year cycle of solar activity. It is noteworthy that a maximum as early as 2:00 A.M. in 1954, has never before been observed.

Sarabhai *et al.* (67) have extended their earlier analysis of changes at the Carnegie Institution stations by examining further unpublished data from 1946 to 1953 supplied by Forbush. They find that while changes of amplitude at the different stations are not correlated in the new solar cycle, the changes

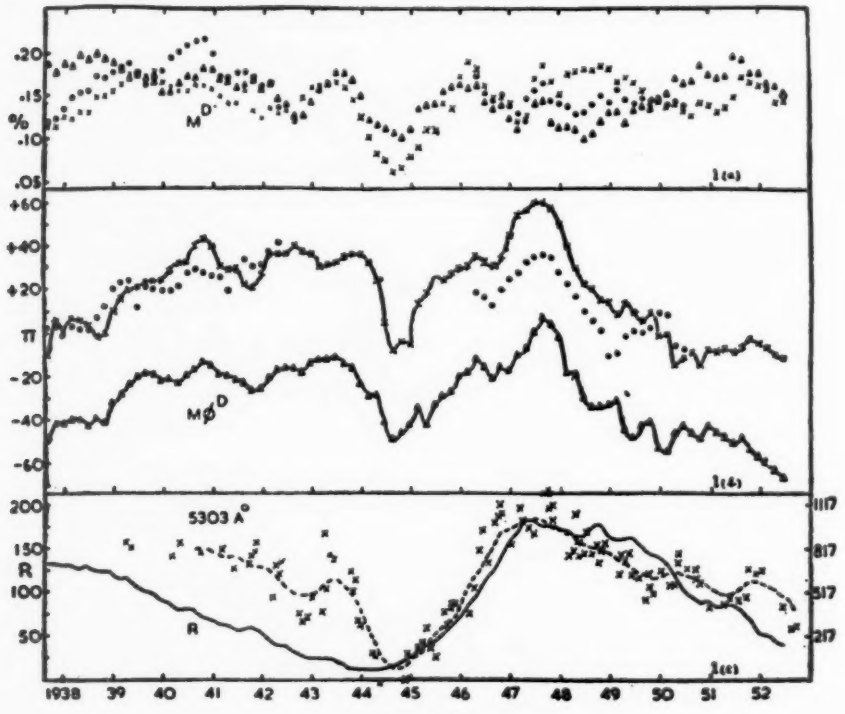

FIG. 8A

of time of maximum continue to exhibit worldwide characteristics. Furthermore they are better correlated with activity of 5303 Å coronal emission than with the relative sunspot number or the magnetic character figure.

The shift of time of maximum of the diurnal component has been shown

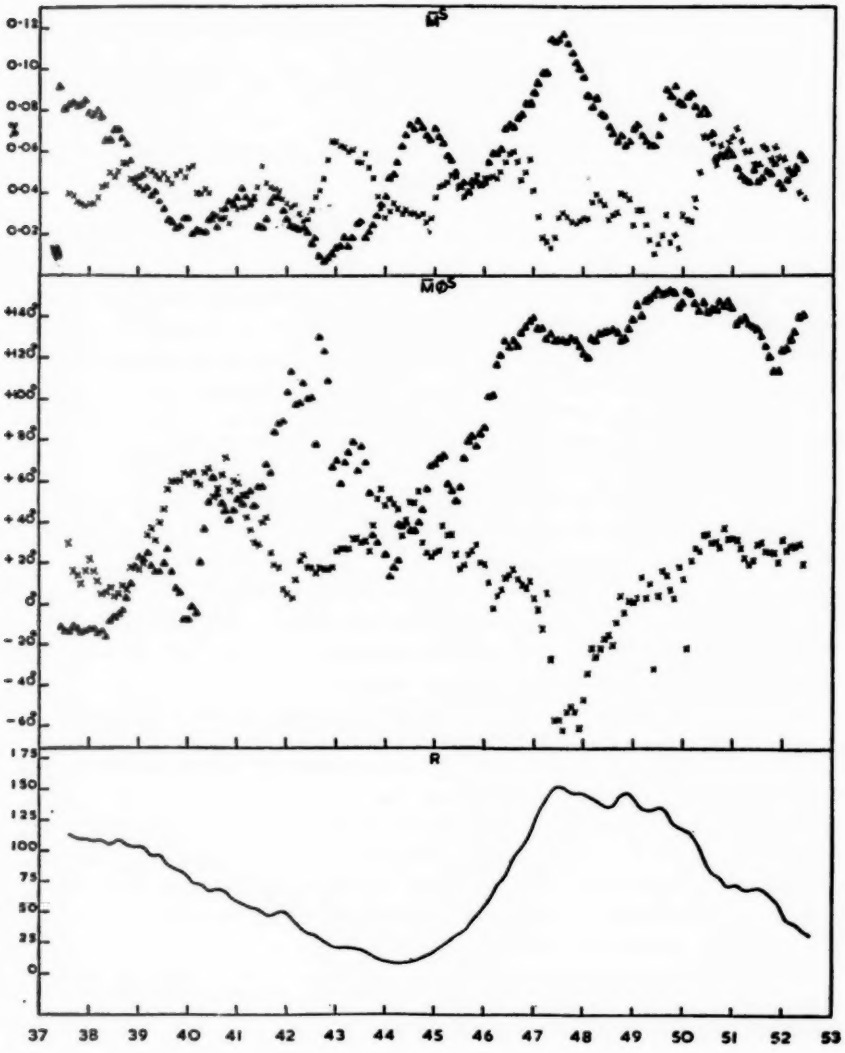

FIG. 8B

FIG. 8. Long term changes of diurnal component. (Fig. A, upper) and semidiurnal component (Fig. b, lower) of the daily variation at Huancayo (▲), Cheltenham (×) and Christchurch (●). M^D and M^S represent amplitude and $M\phi^D$ and $M\phi^S$ the times of maxima of the two components respectively [Sarabhai, Desai & Venkatesan (4, 67)].

by Sandstrom (68) to be independent of the direction of arrival of the particles and of the magnetic disturbance of the days characterised by K_p indices. Sarabhai *et al.* (4) have compared the changes in time of maximum of the 12 monthly mean daily variation studied simultaneously by an ion chamber at Huancayo and a vertical counter telescope at Ahmedabad. For these stations in low latitudes, the changes are well correlated for the short period considered, but the magnitude of the change with the counter telescope is about 2.5 times greater than the change with the ion chamber. Thus the semiangle of the detecting instrument influences not only the amplitude of the daily variation but the long term changes observed in the variation.

Sarabhai *et al.* (4) have demonstrated that in addition to long term changes in the diurnal component of the daily variation, significant changes also take place in the semidiurnal component. The changes occurring at the Carnegie Institution stations are shown in Figures 8(A) and 8(B) which relate to the diurnal and the semidiurnal components respectively. When the daily variation is considered as a whole instead of in terms of its harmonic components, a very remarkable sequence of changes is revealed as shown in Figure 9. At Huancayo, the 12 month mean daily variation which exhibits one maximum near noon from 1937 to 1943, changes over by 1952 to a variation exhibiting one maximum early in the morning. In the intervening period, there is clear evidence of the progressive increase of the early morning maximum followed by a decrease of the noon maximum. During the period 1945 to 1950 the daily variation therefore exhibits two maxima instead of one. Sarabhai and co-workers thus conclude that to follow the physical process of change of the daily variation, it is clearly necessary to discard individual consideration of only the diurnal component of the variation. The most appropriate manner of studying the changes is by examination of the daily variation unresolved into its harmonic components. As shown in Figure 9, only half the cycle of change appears to be completed in 11 years. This is again indicative of a 22 year cycle of change of the anisotropy.

Sarabhai *et al.* (4) have suggested that changes of the daily variation could be considered in terms of the contributions of two distinct types of variations. Both have one principal maximum. For the 'd' contribution the maximum occurs near noon and for the 'n' contribution at about 0300 hours. It has been shown by them that the pattern of addition or subtraction of the day and night contributions of the daily variation is worldwide in character and can satisfactorily explain the observed changes from year to year at Huancayo as well as at Cheltenham. The activity of the two contributions is closely related to the solar cycle of activity. In general, the day and the night contributions are simultaneously added or attenuated. However, just preceding sunspot minimum, there is a brief period when only the day contribution appears to be active. This is immediately followed by an equally brief period when only the night contribution is active. The pattern of addition and attenuation of the contributions appears to be reversed after 11 years.

3.2 Short Term Changes

3.21 *Magnetic storm type changes.*—Large worldwide decreases of cosmic ray intensity associated with magnetic storms have been reported in the past. These decreases have several remarkable features. All magnetic storms are not associated with decrease of cosmic ray intensity, and when effective storms take place, the ratio of change of magnetic field strength to the change of cosmic ray intensity is not constant from event to event. In storms such as the large storm on March 1, 1942, there is almost no latitude

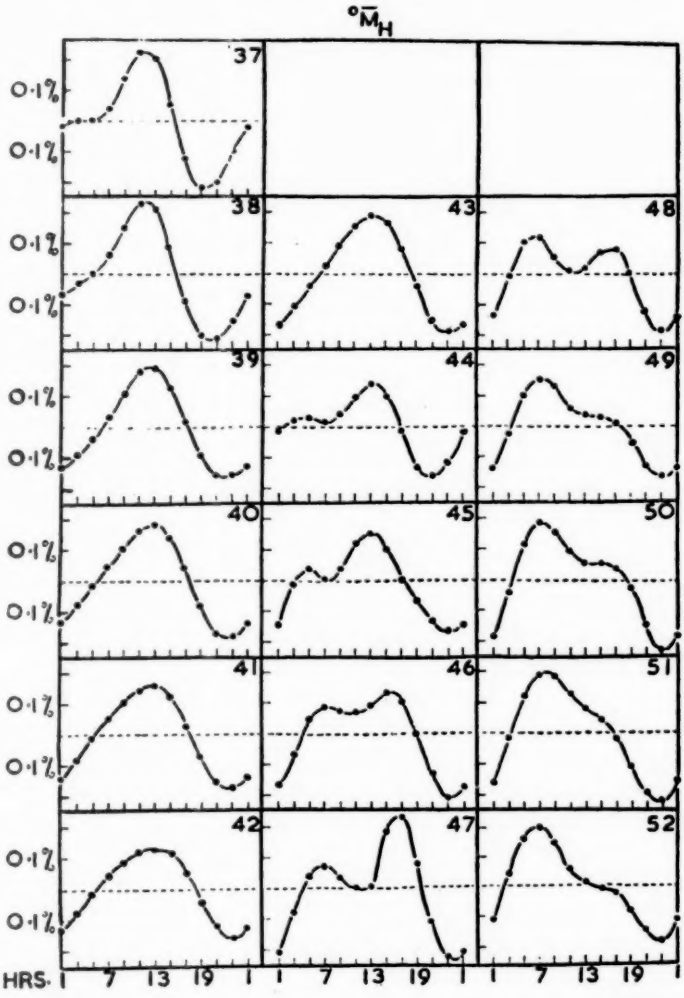

Fig. 9. The 12-month mean daily variation of meson intensity at Huancayo during different years [Sarabhai, Desai & Venkatesan (4)].

effect observable in the magnitude of the decrease at different stations extending from the equator to high latitudes. On the other hand in some storms, the low energy component of the primary cosmic radiation appears to be disturbed more than the high energy component. Neher & Forbush (70) have compared a storm type decrease observed at 70,000 feet over Texas with the decrease in the ion chamber at Mt. Wilson and have showed that the effect was about four times more pronounced in the former case.

During the past three years, further work has been done in studying the solar and terrestrial relationships of magnetic storm type decreases in cosmic ray intensity. Chasson (71) has shown that decreases of intensity occurred on two occasions an appreciable time before measurable geomagnetic disturbances. Trefall (72) has concluded that magnetic storms with a dominent positive peak and a negative peak which is small or absent, produce no change of intensity. Trumpy (73) and Kitamura (74) have examined the characteristics of some magnetic storms. Sekido *et al.* (75) have studied the solar relationship of cosmic ray effective and noneffective magnetic storms, which they designate as S and M type storms respectively. They have found that the frequency of S type storms varies in step with the 11 year cycle of solar activity, but this is not the case with M type storms which they associate with the supposed M regions on the sun. There is correlation between the occurrence of S type storms and the central meridian passage of large sunspot groups. This feature is absent for M type storms. The 27 day recurrence tendency as seen with Chree diagrams, shows sharp peaks for M type storms but sinusoidal changes for S type storms. The authors conclude that S type storms are caused by wide corpuscular clouds ejected from sunspot groups, but the M type storms are caused by narrow corpuscular beams from M regions.

3.22 *Day-to-day changes of intensity.*—The day to day changes of intensity, some of which are of the 27 day recurring type, have been extensively studied in recent years. Early work related to small changes noticed in ionisation chambers, but a significant step forward was possible with data from Simpson's neutron monitors in which it was quite obvious that large and significant changes of intensity of a few per cent in amplitude were occurring almost continuously from day to day. Simpson *et al.* (76) showed the association of the increases with the central meridian passage of active solar regions, and particularly of regions of green coronal emission on the sun. These increases were often seen to be followed within two to three days by increased geomagnetic activity. Recently Simpson *et al.* (77) have shown the close association of the central meridian passage of unipolar UM regions with cosmic ray increases during 1953. These regions as well as the increases of intensity persisted for several solar rotations.

Several workers have now demonstrated that the day to day changes are worldwide in character. Fonger (17) has found that changes in the neutron monitor at Climax are similar to simultaneous changes observed in the ion chamber at Freiburg, but on the average are five times greater in

amplitude. Neher & Forbush (70) have demonstrated the correlated changes occurring in the ion chambers at Huancayo and Cheltenham, the neutron monitor at Climax and the ionisation at 70,000 feet over Bismarck. These are shown in Figure 10 which reveals the difference in amplitude of changes in

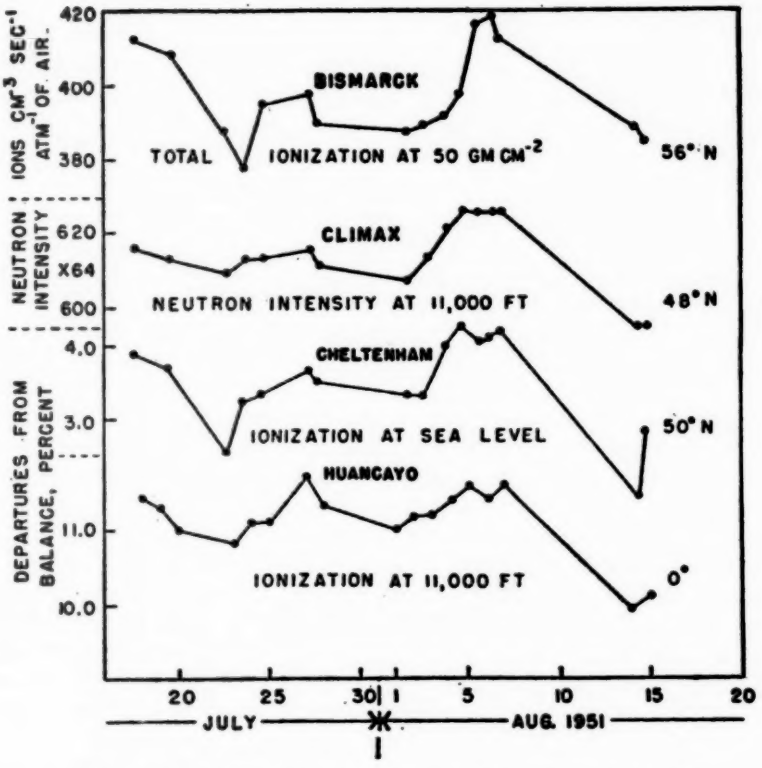

FIG. 10. Correlated day-to-day changes of cosmic ray intensity [Neher & Forbush (70)].

each case. Neher *et al.* (60) have demonstrated that the changes at high altitude over Bismarck and near the north geomagnetic pole are correlated amongst themselves and with changes in an ion chamber at ground level. van Heerdan & Thambyahpillai (35) have recently investigated the correlated changes measured with a neutron monitor and a counter telescope at London. In the study of the 27 day recurrence of these changes, they find a decrease of mean intensity connected with the increase of amplitude of the fluctuations. They have thus come to the conclusion that the changes represent decreases of intensity. Meyer & Simpson (78) have studied the 27 day recurrence tendency in the ion chamber data of Huancayo and the neutron

monitor at Climax, and have demonstrated that the recurrence tendency is altered during the solar cycle. It is more pronounced during sunspot maxima, when incidentally the general intensity is low, and is weak at sunspot minimum when the general intensity is maximum.

Meredith *et al.* (63) have made rocket measurements with a single counter capable of responding to radiation of intensity as low as 20 Mev. They find that on many days during 1954 there is, at an altitude above 40 km, a large intensity of radiation of energy between 20 Mev and 140 Mev. It is of course debatable whether radiation of this type can legitimately be classed in cosmic radiation. But the observations show that just like the day to day variations of high energy radiation, there is also a day to day variation of large magnitude occurring in much softer radiation which can only be observed at very high altitudes.

3.23 *Day-to-day changes of anisotropy.*—The change of the diurnal component of the daily variation of cosmic rays during magnetically disturbed days has been extensively studied (39, 68, 73, 79 to 83). It is found that on days with high K_p, the amplitude of the diurnal component increases and its time of maximum becomes earlier. Sekido & Kodama (81) have shown that this effect is present to a greater degree in measurements made with narrow angle telescopes than with instruments with wider directional sensitivity. Sekido & Yoshida (84) have moreover demonstrated that the semidiurnal component which is normally present to a greater extent in the daily variation measured with narrow angle telescopes than with telescopes of large semiangle, gets reduced on magnetically disturbed days.

Sekido *et al.* (85) have made an intensive study of the mean storm type vector representing the difference in the diurnal components of the disturbed period daily variation and the quiet period daily variation during each year. They have shown that the disturbance vector representing a storm type anisotropy points on the average towards the radially outwards direction from the sun, and undergoes a long period change from year to year, broadly in step with the 11 year cycle of solar activity.

Following the discovery of long term changes in the anisotropy of the primary radiation as revealed by changes in the 12 month mean daily variation of cosmic ray intensity, various attempts have been made to study the short term changes of the anisotropy, not necessarily connected with magnetic disturbances. Firor *et al.* (79) have pointed out that the daily variation of the nucleonic component as revealed by the neutron monitor is highly variable in character. They found groups of days on which there was a large diurnal variation with a maximum during the day. There were other days on which there was no appreciable daily variation, and indeed on others, the daily variation appeared to have a maximum in the night. Sittkus (86) has looked at the amplitude of the diurnal variation measured by an ion chamber on individual days and he has noticed the tendency for a variation of large amplitude with a maximum near noon to occur during groups of days. He has found a 27 day recurrence tendency for these groups but com-

pared to the rest of the days, the days with large noon time amplitude are not associated with a significantly different magnetic character figure. Remy & Sittkus (87) have shown that during 1954, the occurrence of days on which there was a large day time maximum of intensity was rarer than in 1953. They have moreover noticed a great variability of the time of maximum of the diurnal component of the daily variation.

Weighty evidence concerning the change of anisotropy from day to day comes from the investigations with narrow angle telescopes reported by Sarabhai & Nerurkar (88). With telescopes having semiangles of 2.5° and of 5° in the E-W plane, the combined data for each type of telescope for every day were examined.

On about 75 per cent of the total days the daily variation exhibited only one maximum. For these days they have found, as shown in Figure 11, a marked tendency for the diurnal maximum during the year 1954 to occur either at 12:00 noon or in the early morning at about 0300 hours local time. On the basis of a phenomenological classification of days according to whether they exhibit a maximum during the day, a maximum during the night or they exhibit two maxima instead of one, the days have been desig-

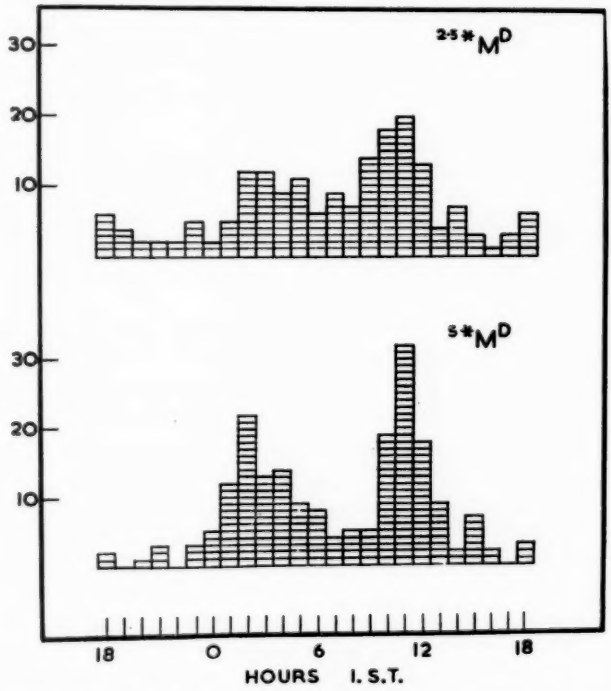

FIG. 11. The distribution of occurrence of the diurnal maxima for each of 24 hours of a day. The upper and the lower histograms refer to telescopes of 2.5° and 5° semiangles respectively in the E-W plane [Sarabhai & Nerurkar (88)].

nated as 'd', 'n' & 's' respectively. The three types of daily variations obtained for data averaged for days of each type are shown in Figure 12. The results indicate that there are at least two preferred orientations of the anisotropy and that on any particular day which does not belong to the 's'

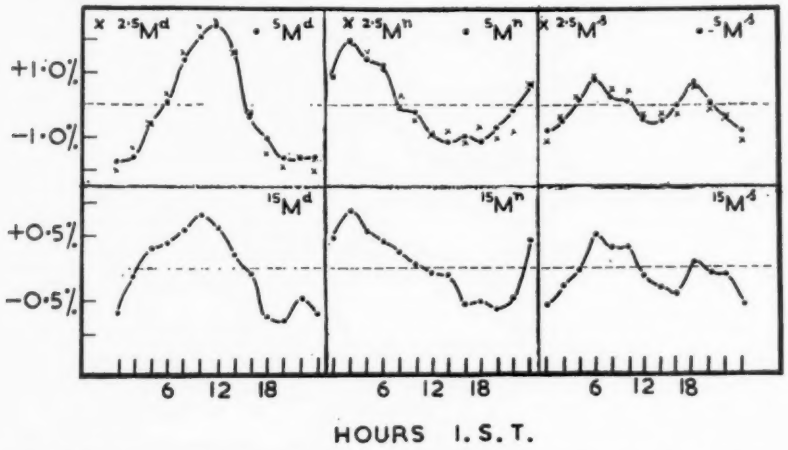

FIG. 12. The average daily variation of meson intensity measured with telescopes of 2.5°, 5° and 15° semiangles on 'd', 'n' and 's' type days [Sarabhai & Nerurkar (88)].

type, there is present either one of these two types of orientations. The amplitude of the daily variation occurring on each of these types of days is large and is of the order of 1.5 per cent to 2.0 per cent as observed with a narrow angle telescope at low latitudes. These days, particularly of the 'd' type, have a 27 day recurrence tendency and usually occur in groups as observed by Sittkus.

The recurrence tendency of the anisotropy observed in the ion chamber data at Huancayo has been studied by Yoshida & Kondo (89). They found a strong recurrence tendency, extending to two solar rotations, which was more marked than the recurrence tendency in either the general cosmic ray intensity or in the magnetic character figure during the same period. Kane (90) has studied the recurrence tendency in the daily variation measured by neutron monitors at several stations and by the Freiburg ion chamber for the years 1951, 1952, and 1953. He has shown that the recurrence tendency in the daily variation, unlike the tendency in the general intensity, remains almost unaltered during the three years.

3.3 Solar Flare Effect

Since the detailed review by Elliot (1) no major solar flare effect was observed in cosmic radiation until recently on February 23, 1956. In the four early events large increase of intensity was seen to occur simultaneously in instruments at stations situated in latitudes higher than 25°. The in-

creases lasted from one to three hours and in three cases followed the visual observation of the flare within an hour. They were larger in neutron detectors than in instruments measuring meson intensity and the absence of recorded increases at low latitudes suggested that the radiation responsible for them was much more strongly energy dependent than normal cosmic ray intensity. There was no evidence for increases in the rate of extensive air showers (91).

During the last three years much further examination and analysis of earlier data has been done. Graham & Forbush (92) have confirmed in the records of an ion chamber at Thule on July 25, 1946 the occurrence of a polar type increase as reported earlier from Resolute and Godhavn. On the assumption that the flare effect is caused by the emission of solar particles of cosmic ray energy, Schluter (8) and Firor (9) have examined the trajectories of charged particles in the geomagnetic field. Figure 13 shows the impact zones of solar particles in the rigidity range two to ten Bev in terms of local time at the place of observation. The impact zone theory, on assum-

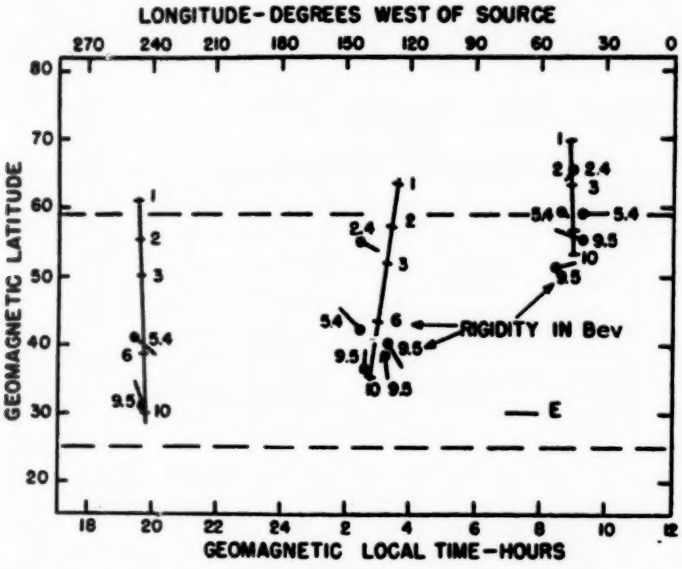
LONGITUDE-DEGREES WEST OF SOURCE
270 240 210 180 150 120 90 60 30 0
80 70 60 50 40 30 20
GEOMAGNETIC LATITUDE
18 20 22 24 2 4 6 8 10 12
GEOMAGNETIC LOCAL TIME-HOURS
RIGIDITY IN Bev
E

FIG. 13. Impact points on the earth for one to ten Bev particles approaching in the equatorial plane from a source situated at noon local time [Firor (9)].

ing a solar diameter of about 30° as viewed from the earth, satisfactorily explains the principle features of the increases observed in middle latitudes. However there is difficulty in explaining polar type of increases at latitudes higher than 65° on the basis of direct orbits from the sun.

Sekido & Murakami (93) have analysed the amplitude and the duration of the solar flare increases observed at different stations during each of the four early events. They distinguish between the effect occurring within the

impact zones from the polar type of effect, which has a longer duration and a less steep rise of intensity than the former. This is suggestive of a different mode of travel of particles for the polar type increases as compared to the direct trajectories for arrivals within the impact zones. These authors suggest that the flare radiation has a spectrum such as E^{-5}. However there is a varying amount of averaging of the solar flare increases at different stations and there is also a difference in the geometry of recorders at various stations. Hence a derivation of any definite power law spectrum at the present moment appears premature, particularly because of the imperfect understanding of the polar type increases.

Treiman (94) has calculated that the solar magnetic moment should not exceed 5×10^{32} gauss-cm.^{3}, if the calculations of the impact zones by Firor are to be retained. Kraushaar (95) has made a similar calculation concerning the galactic magnetic field and puts an upper limit of 3×10^{-6} gauss for it. Kraushaar (96) has offered an interesting suggestion involving deflections in a galactic field of this magnitude and emission of particles from the sun over a wide range of angles for explaining the polar type of increases. A verification of this explanation would be possible from the seasonal dependence of the flare effect at polar latitudes, but it is difficult at present to confirm the validity of his hypothesis from the few instances which have been recorded.

Firor (9) has looked for flare type of increases associated with small flares of magnitude 1^+. As shown in Figure 14, he finds about 1.0 per cent increase in the intensity on days when the Climax recorder was in an appropriate impact zone to be able to receive particles from the sun. This he compares with no increases on days when the Climax recorder was not in one of the impact zones, during the occurrence of flares. Neher *et al.* (60) have reported in the records of a flight made on July 23, 1951, an increased intensity during the descent as compared to the ascent of the instrument. A flare of magnitude 1 occurred about an hour before the commencement of the flight. This is the solitary reported instance when an instrument was aloft soon after a flare, and the authors indicate that in terms of solar protons the increase would have to be due to arrival of 0.6 Bev particles. It is perhaps significant to note that the low energy cut off at the time was about 0.8 Bev for general cosmic ray intensity.

The most recent flare of February 23, 1956 has been remarkable in many respects. It was of magnitude 3^+ and apparently more pronounced cosmic ray increases have been recorded than during the earlier events. An increase of about 4500.0 per cent has been reported in a neutron monitor at latitude 57°N. by Marsden *et al.* (97). Moreover an average increase of 5.7 per cent at stations near the magnetic equator in India is reported by Sarabhai *et al.* (98). On the assumption that the increase is caused by solar protons travelling along more or less direct paths, they suggest from Brunberg & Dattner's calculations that only solar protons in the energy range 30 to 67.5 Bev could

have been responsible for the increases at the Indian stations. These particles could only come from restricted directions inclined to the zenith towards south and hence the small per cent change of intensity is indicative of a large flux of protons of about 50 Bev energy which they estimate is nearly 1.5 times the flux of the background cosmic rays in the same energy range. Thus the upper limit of energy of particles responsible for flare type increases is much higher than has been hitherto believed.

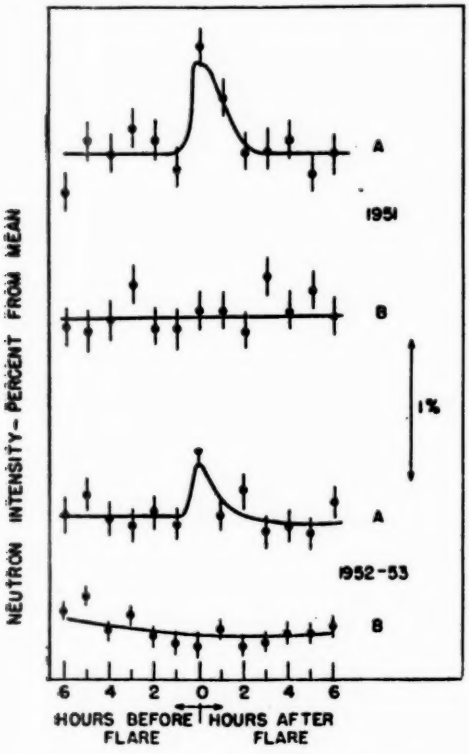

FIG. 14. Cosmic ray intensity during times of small solar flares. Curves A and B show behaviour when detector was within and outside respectively of the morning impact zone [Firor (9)].

4. THE INTERPRETATION OF VARIATIONS

4.1 DEPENDENCE ON MEAN ENERGY OF PRIMARIES

From various experiments which have been referred to in the earlier section, we have attempted to summarise data of significance to the evaluation of the dependence of the variations on the mean energy of primary radiation. These are presented in Table I.

TABLE I

RATIO OF AMPLITUDES OF VARIOUS TYPES OF CHANGES RECORDED BY DIFFERENT RECORDERS*

Type of change	Recorder A	Recorder B	Recorder C	Ratio of amplitude A:B or A:B:C	Ref.
Long term change	IC 50°N. sea level	IC 1°S. 11,000 ft.	IC 43°S. sea level	1:1:1	(3)
	IC 50°N. 90,000 ft.	IC 1°S. 11,000 ft.		12:1	(56)
Magnetic storm type change	NM 48°N. 11,000 ft.	IC 49°N. sea level		5:1	(100)
	NM 42°N.	IC 1°S. 11,000 ft.		2.7:1	(3)
	IC 1°S. 11,000 ft.	IC 50°N. sea level	IC 80°N. sea level	1:1:1	(91)
	CT(±12°) 25°N.	CT(±40°) 25°N.	CT(±85°) 25°N.	3:1.7:1	(40)
	CT(±45°) 47°N.	IC 49°N.		1.5:1 to 2.4:1	(38)
	IC 70,000 ft.	IC sea level		4:1	(70)
Day-to-day change	NM 48°N. 11,000 ft.	IC 49°N. sea level		5:1	(17)
	IC 1°S. 11,000 ft.	IC 50°N. sea level		1:1	(70)
	IC 50°N. 70,000 ft.	NM 48°N. 11,000 ft.	IC 1°S. 11,000 ft.	7:3:1	(70)
Diurnal variation	NM 48°N. 11,000 ft.	NM 1°S. 11,000 ft.		1:1	(79)

* The recorders are designated by IC for ionization chamber, CT(±x°) for counter telescope of semiangle x° in the E-W plane and NM for neutron monitors. The geomagnetic latitude and the elevation of the observing station are indicated by numbers immediately following the abbreviation used for recorders.

TABLE I—(*Continued*)

Type of change	Recorder A	Recorder B	Recorder C	ratio of amplitude A:B or A:B:C	Ref.
	IC 1°S. 11,000 ft.	IC 50°N. sea level		1.1:1	(42)
	CT(±45°) 47°N.	IC 49°N.		1.5:1 to 2.4:1	(38)
	CT(±12°) 25°N.	CT(±40°) 25°N.		1.3:1	(40)
	CT(± 5°) 13°N.	CT(±15°) 13°N.		2:1	(41)

It will be noticed that the long term changes of intensity, the magnetic storm type decreases and the day to day changes of intensity exhibit certain common features. The amplitude of variations studied with omnidirectional instruments is generally smaller than with counter telescopes where the amplitude becomes progressively larger with a dimunition of angle of opening of the telescope. There is often a larger change of intensity of particles of energy less than 10 Bev than of those above it. The ratio, however, of the changes observed in the two cases is variable. Moreover, there are occasions when, as shown by Neher, the reverse is true, and there can be increase of intensity at intermediate energies without corresponding change at low energies. It is remarkable that the fluctuations are not as latitude dependent as a comparison of the neutron monitor studies with charged particle detectors at the same latitude would seem to suggest. It appears that the difference between the semiangles of the meson detector, which is usually an ion chamber or a wide angle telescope, and the effective directional sensitivity of the neutron monitor, is perhaps responsible for this discrepancy. It has been discussed earlier that the energy spectrum of solar primaries associated with flares is considerably steeper than the spectrum of normal cosmic ray primaries.

The amplitude of the diurnal component of the daily variation of intensity has frequently been considered an index of the degree of anisotropy of primary radiation. However, the amplitude depends very much on the directional sensitivity of the measuring instrument and on the period for which data are averaged. At the present moment we have no available data where the amplitude of the daily variation observed with a neutron monitor can be compared directly with the amplitude of the daily variation in meson intensity measured with a telescope of equivalent effective directional sensi-

tivity. Until this is done, it would be difficult to conclude whether the anisotropy is greater in respect of the spectrum in the mean energy range of about 7 Bev which is measured in a neutron monitor at middle latitudes and in the spectrum with a mean energy of about 40 Bev which would be measured at a similar station with a meson intensity recorder.

Some evidence on the energy dependence of the anisotropy is obtainable from the latitude effect of the amplitude of the daily variation observed with similar instruments. Huancayo is at a mountain elevation at the geomagnetic equator compared to Cheltenham which is at sea level at 50°N. geomagnetic latitude. Over a period of several years, the per cent amplitude at Huancayo is about ten per cent larger than at Cheltenham. Between Huancayo and Climax there is no appreciable difference in the amplitude of the neutron daily variation over 13 months. Thus within the latitude sensitive spectrum of primaries there appears to be no marked change in anisotropy. Experimental data for high energy primaries from experiments made with large atmospheric showers or at great depths underground establish only an upper limit to the magnitude of the anisotropy at these energies and in consequence do not reveal the dependence of the anisotropy on primary energy.

4.2 Solar and Terrestrial Relationships of Cosmic Rays

There is now a vast amount of evidence to show that the sun strongly influences the cosmic rays that are measured on the earth. Most of these influences have been revealed through relationships with the activity of the sun which changes markedly from time to time. Some activity persists through several solar rotations and produces characteristic 27 day recurrences. General solar activity changes with an 11 year period, while the state of magnetic polarity of bipolar sunspots has a periodicity of 22 years.

The sun appears to influence the low energy cut off of the primary spectrum over an 11 year cycle. Independently of the change of cut off, it is known to also produce a general change of intensity, with a cycle of 11 years. It produces changes of intensity of shorter duration. Many of these are related to the central meridian passage of regions of solar activity such as sunspot groups, unipolar regions and regions of intense coronal 5303 Å emission. The occurrence of the regions at low heliographic latitudes appears to favour their effectiveness in influencing cosmic rays.

The sun produces an anisotropy of the cosmic radiation. The 12-monthly average anisotropy illustrates a change which is consistent with a cycle of 22 years and perhaps constitutes the first solar influence detected so far on the earth which has a period corresponding to what may be considered the true solar cycle of activity. This requires to be verified by observations extending over several cycles. The sun also produces day to day changes of anisotropy, some of which are related to the central meridian passage of active regions as in the case of variations of intensity.

The sun emits cosmic rays in association with flares, and these are now

believed at least sometimes to have particles of energy exceeding 50 Bev, if they are protons which have travelled in direct paths to the earth.

The terrestrial relationships of cosmic ray variations with geomagnetic activity arise from the latter being themselves associated with solar activity. Similar is the case with some ionospheric disturbances. For instance Dolbear *et al.* (99) have shown the association of cosmic ray increases with ionospheric disturbances during day time hours. Also flare type increases of cosmic rays are associated with ionospheric disturbances and radio fade outs.

4.3 The Mechanisms of Variations

The close relationship of cosmic ray variations with solar activity has prompted various workers to investigate whether these changes are caused by the emission of cosmic rays from the sun. At the present moment there is general agreement on solar emission only in respect of the flare type increases. The view is tending towards the modulation of cosmic ray intensity to explain the other variations because of the combined weight of one or more of the following arguments which by themselves do not warrant an unequivocal answer.

(*a*) At minimum of solar activity, as judged by visual evidence on the solar disc or by comparative absence of terrestrial effects such as magnetic and ionospheric disturbances and auroral activity, there is an absence of "knee" with the presence of many low energy particles with energy down to about 150 Mev. Associated with these particles, there is an increase in general intensity, not only because of the absence of the low energy cut off but, due to enhanced intensity, throughout the whole spectrum. This result suggests that turbulent magnetised clouds produce a screening of the earth probably by scattering of cosmic rays. The characteristics of the screen are governed by the solar activity. At sunspot minimum the screen is either temporarily removed or is made less impervious.

(*b*) Magnetic storms on many occasions are accompanied by decreases in cosmic ray intensity, but never with increases. Heerdan & Thambyahpillai (35) have shown that an increase in the general intensity is associated with the reduction of amplitude of 27 day recurrences, thus suggesting that the 27 day recurrent changes are decreases. They are therefore similar to the magnetic storm type changes and indicate that screening of cosmic ray intensity has occurred. This is in general agreement with the minimum of amplitude of 27 day recurrences occurring at minimum solar activity when there is general enhanced total intensity.

The only evidence for an increase in cosmic ray intensity associated with visible solar disturbances is that put forward by Simpson *et al.* (76, 77). Figure 15 illustrates the sharp peaks of intensity which coincide with the central meridian passage of active regions. It is not clear at the present moment that there is any reasonable argument to minimise the significance of this type of observation. Therefore there is at the present moment a real difficulty in assuming that all variations under consideration here are in the

nature of decreases. More data on the association of increases with the central meridian passage of regions of solar activity are required to settle this point.

Because of close relationships with geomagnetic disturbances, early workers tried to explain the variations in terms of a modulation of cosmic ray intensity by changes in the geomagnetic field. Simpson (100) has shown that the variations in the neutron intensity at latitudes above the "knee"

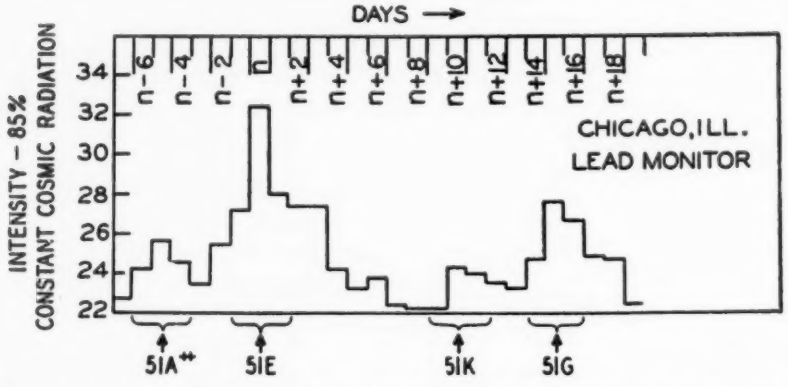

FIG. 15. Association between intensity maxima in neutron pile at Chicago and the central meridian passage of local active regions on the sun. Simpson, Fonger & Wilcox (76).

of the intensity versus latitude curve were very similar to those observed at low and middle lattitudes. Hence these variations in the nucleonic and the ionizing component are not produced by geomagnetic field disturbances. Nagashima (18) has proposed a static electric field surrounding the earth and assumes changes in it to explain the worldwide nature of the intensity variations. His model explains fairly well the altitude dependence of the variations reported by Neher *et al.* (60) and by Fonger (17), as well as their latitude dependence examined by Yoshida & Kamiya (101). However, Simpson (100) has examined the effect on cosmic ray intensity of axially symmetric geoelectric fields at different distances from the earth. He finds that in the range 45°N. to 60°N., the latitude dependence of the fractional changes in neutron intensity during different periods does not agree with the latitude effect calculated for different distances of the field. He concludes that no accelerating or decelerating geoelectric field is responsible for these variations. In addition, such a mechanism will be unable to account for large cosmic ray changes during periods when there are no geomagnetic disturbances and presumably no perturbing geoelectric fields. Simpson therefore suggests that the mechanism responsible for the changes is of extraterrestrial origin and its association with solar phenomena is independent of the earth's system.

Alfvén (102) has suggested that the emission of corpuscular matter from the sun which is responsible for magnetic storms is also responsible for cosmic ray fluctuations. An electric field is set up due to the polarisation of the neutral beam which takes place in the presence of a trapped magnetic field carried with it. The acceleration and deceleration of cosmic rays traversing the field may be expected to cause increases or decreases. This model has been considered in greater detail by Brunberg & Dattner (103), to explain the decreases observed with magnetic storms. They consider the orbits of cosmic rays in a general dipole field in the solar system and require a magnetic field of 10^{-5} gauss in the vicinity of the earth, if only decreases are to be observed.

Davis (104) has discussed the influence of the emission of ionized matter from the sun on solar or galactic magnetic fields within the solar system. He explains that the tendency would be to sweep the magnetic fields away and leave a field free cavity of a mean radius of about 200 astronomical units around the sun. Cosmic rays of rigidity less than 10^{12} volts would be trapped within the cavity and their intensity would depend on its volume. He has tried to show that a 1.0 per cent change of the mean radius of such a cavity with the 11 year solar cycle could explain the 4.0 per cent change of intensity as observed by Forbush (3). However this mechanism fails to explain the much larger long term changes of intensity for low energy than for high energy primaries, as observed by Neher (55) and by Meyer & Simpson (57).

Morrison (105) has recently proposed a model which attempts to explain most of the principal features of various types of fluctuations observed in cosmic ray intensity which he believes to be decreases of intensity. He supposes that clouds of ionized matter are emitted more or less continuously from active regions of the sun. These clouds have within them turbulent magnetic fields instead of trapped coherent magnetic fields visualised by Alfvén (102). The clouds would be free of cosmic ray intensity when they are emitted, but intensity will slowly build up in them as cosmic rays diffuse into them. The intensity of cosmic rays at any time within the cloud, would be governed by the age of the cloud and the rate of diffusion of cosmic rays into it. The diffusion time depends on the scale of turbulence and the strength of the magnetic field and is therefore greater for low energy particles than for high energy particles. As the earth gets enveloped in one of these clouds, it experiences decreases of cosmic rays intensity depending on the intensity within the cloud at that moment.

Morrison expects that at great distances from the sun of the order of 100 astronomical units, there would be a fairly uniform diffuse cloud in place of the discrete structures seen at closer distances of the sun equivalent to the distance of the earth. Scattering in the distant diffuse clouds is expected to produce the general change of intensity as well as the change in the low energy cut-off accompanying the solar cycle. This scattering would be more effective for low energy particles than for high energy particles and thus explains the experimental observations in this regard.

Morrison has suggested the possibility of being able to explain a cosmic

ray intensity increase associated with the central meridian passage of a UM region by postulating the presence of streams on either side of the UM region which would have more turbulence and therefore less intensity than the stream from the UM region. However not all cosmic ray increases have been associated with UM regions, and there might be difficulty in explaining the day to day increases on Morrison's model.

An attractive feature of Morrison's theory is that turbulent chaos and not a highly ordered model is called upon to explain the observed facts. Morrison thus comments on his model "it lacks the specific details of other proposals, but this weakness perhaps allows better experimental agreement." Morrison does not explain the wide variability of the energy dependence of different variations on different occasions. Perhaps another drawback with the model is that it fails to explain solar anisotropy of radiation revealed by the diurnal variation and its changes.

5. THE INTERPRETATION OF THE SOLAR ANISOTROPY OF COSMIC RADIATION

Explanations of the daily variations of cosmic rays in terms of a solar magnetic field or the variations of a geomagnetic field became untenable quite early, because of the failure to detect the expected large latitude dependence of the variations. Brunberg & Dattner (106) have pointed out that seen from a fixed coordinate system, the rotating sun would be strongly polarised so that there would be a voltage difference between the poles and the solar equator of the order of 10^9 volts. The combined action of the electric field produced by polarisation and the solar magnetic field will make charged particles within the solar system partake in a general rotation with the sun so that the earth will receive an excess of particles in the 18 hourly direction. They thus get a tangential anisotropy. Alfvén (107) on the other hand, has shown the possibility of an outward radial flow of energy depending on the accelerating processes within the solar system. This will impart a radial anisotropy to cosmic rays, which will change with the 11 year cycle of solar activity. These explanations require a general magnetic field of the sun having an intensity of about 10^{-5} gauss at the earth's orbit to account for a diurnal variation whose amplitude was believed to be about 0.2 per cent.

Nagashima (108) has recently examined the possibility of explaining a diurnal variation of the order of 0.2 to 0.5 per cent on the basis of the electric field theory suggested by him to explain the magnetic storm type change of total intensity. There would however be a large latitude dependence of the daily variation which is not observed.

Because of the neglect of the following experimentally determined features of the daily variation averaged over periods of one year or longer, it is difficult to attach much importance to any of the above mentioned theories to explain the observed facts.

(*a*) The daily variation cannot at all times be satisfactorily described

in terms of the diurnal component only. For several years in the past the daily variation had two significant maxima.

(*b*) A shift of the principal maximum of the daily variation by as much as ten to twelve hours takes place over a period of years. The implication of this shift is that during some years, the daily variation exhibits a principal maximum during the day while in others, the maximum is shifted to the night.

(*c*) The amplitude of the daily variation is markedly determined by the directional sensitivity of the recording instrument. The change of amplitude of the daily variation with change of semiangle of the telescope is very much more than can be normally expected. Large amplitudes of the order of 1.2 per cent have been observed with narrow angle telescopes at low latitudes.

The occurrence of the storm type anisotropy and the frequent day to day variations of anisotropy make it necessary to evaluate the physical meaning to be attached to the anisotropy derived from data averaged over a long period. The daily variations of the 'd' and the 'n' type, which occur on groups of days, have times of maxima which coincide with those of the two contributions which are found to play a determining role in producing long term changes in the 12 month mean daily variation. This raises the question whether there is in fact a permanent anisotropy in addition to the variable types of anisotropy present on groups of days. If we accept the existence of a permanent anisotropy, its features and characteristics are at the present moment not known. It would appear that these have been masked by a more prominent anisotropy which is of a highly variable character. At the present moment, no satisfactory place can be found to fit in the above picture the semidiurnal type of variation observed on certain days at low latitudes.

At the present juncture it is important to find explanations for the two main types of variable anisotropies. In this connection theories which have been advanced by various workers to explain the characteristics of the storm type anisotropy assume a special significance. Elliot & Dolbear (83) and Alfvén (102) have suggested that electric fields are produced within beams of ionized particles emitted from the sun in the presence of the solar magnetic field. Acceleration of cosmic ray particles traversing these beams produces an anisotropy in particular directions depending on the relative position of the beam and the earth. Nagashima (108) has examined the detailed implications of this model taking into consideration the deflection within the beam of particles in the trapped magnetic field which is derived from the solar dipole field. A feature of his theory is that associated with every beam there is a minimum energy below which no anisotropy would be produced. For higher energies, the per cent anisotropy would progressively decrease with increasing energy. Nagashima has shown that the maximum anisotropy would be produced in the direction pointing towards the sun. This agrees with the experimental determination of the storm type vector by Sekido *et al.* (85). Nagashima's theory also explains the 11 year cycle of variation

of the storm type anisotropy as being due to a change in the density of the solar streams and of the strength of the trapped magnetic fields during the solar cycle of activity.

Nagashima's theory does not explain a night time maximum as observed in the daily variation on groups of days. Recently Nerurkar (109) has extended Nagashima's work by admitting the possibility of random orientation of the magnetic field trapped within the beam. This could arise if the orientation of the field in the beam is not determined by the solar dipole field but by the field in the direct vicinity of the regions from which the beams are emitted. Since the daily variation reveals an anisotropy in the E-W plane, the component of the trapped field perpendicular to the equatorial plane has to be considered. Thus a modification of Nagashima's theory whereby the direction of the effective magnetic field can be reversed is possible. Nerurkar has demonstrated that there can then be anisotropy in the 0800 and in the 1600 hour directions depending on the orientation of the magnetic field. Due to the effect of the geomagnetic field these anisotropies can produce at low latitudes maxima of intensity in the daily variation at 0300 and 1100 hours respectively in agreement with observations. The most satisfactory scheme of things would be, to be able to explain intensity fluctuations as well as anisotropy on a unified theory. Further work is now required in this direction.

6. IMPLICATIONS ON THEORIES OF THE ORIGIN OF COSMIC RAYS

The presence of heavy nuclei and the isotropy of the primary radiation have a direct bearing on our concepts of the origin of cosmic radiation and of the properties of interplanetary and interstellar space. The composition of the primary radiation in terms of the relative abundance of nuclei with different atomic numbers sets an upper limit to the number of collisions, and to the mean life time of the primaries. This, coupled with knowledge of the density of matter in interstellar space gives an estimate of the volume in which the radiation could be confined. The containing of the cosmic rays within a specified volume is required from energy considerations of the universe as a whole, as also from the necessity to produce a high degree of isotropy through a state of equilibrium reached within this volume.

As we have discussed earlier, the existence of a permanent solar anisotropy may be considered to be uncertain at the present moment. If it is present, it is not likely to cause an amplitude of the daily variation larger than 0.5 per cent. A solar anisotropy of a highly variable character which may on occasions have large magnitude of the order of one or two per cent does exist, but this appears to be caused by the modulation of particles in the neighbourhood of the earth in their passage through streams of ionized particles emitted from the sun with magnetic fields trapped within them.

There is some evidence for a galactic anisotropy pointing to an excess of particles coming from the direction of the milky way, and particularly from the galactic centre. Davis (110) has discussed the implications of these

results in terms of the galactic magnetic field along the spiral arm in which the solar system is situated, and in terms of various mechanisms proposed for the acceleration of particles to cosmic ray energies. He calculates the anisotropy to be expected through three processes in the very high energy primaries as studied in extensive air shower experiments. He comes to the conclusion that excepting for the results of Daudin & Daudin (51), the observations of Cranshaw & Galbraith (52) and of Farley & Storey (44) favour acceleration by Fermi's mechanism and the diffusion of cosmic rays along the spiral arm.

A theory of variations and of origin of cosmic rays within the solar system has been proposed by Alfvén (107). This requires the presence of a permanent solar field of the order of 10^{-5} gauss at the earth. Particles get accelerated to cosmic ray energy by repeated traversals through disturbed electromagnetic fields produced in the solar system by storm producing beams. By circulation in the trapping field the radiation would become almost isotropic up to 10^{14} ev. The absence of anisotropy of any large magnitude for primaries of 10^{16} ev energy as shown by experiments with extensive air showers would be contrary to the predictions of this theory.

Evidence for the stellar origin of cosmic rays comes from the solar flare effect and from the discovery of a point source reported by Sekido *et al.* (54). Firor *et al.* (111) have estimated from the frequency of the occurrence production of 4 Bev protons at the rate of 10^{23} particles per second. Sarabhai *et al.* (98) have recently estimated that at the time of a large flare of intensity 3^{+}, about 10^{28} protons have been accelerated to energies of about 50 Bev. The estimate of Firor *et al.* (111) is compatible with the intensity of cosmic rays in the 4 Bev region if all solar particles are trapped in a region of mean radius of the order of 10^{15} cm. At the present moment, it would appear that stellar bodies could perhaps provide an adequate mechanism for injecting into the galaxy particles which through other accelerating mechanisms of the type suggested by various workers (112 to 116) could contribute to the overall flux of intensity.

7. CONCLUSION

New experimental facts concerning cosmic ray variations and the anisotropy of the radiation have given a great stimulus to theories of origin of cosmic rays and of cosmical electrodynamics. We now accept the existence of large changes of primary intensity. We have good reason to believe that many of the changes represent modulations of primary intensity occurring at some distance from the earth. The changes therefore provide a new and valuable tool to study electromagnetic fields and distribution of matter in interplanetary and interstellar space. There appears a possibility of being able to understand these changes in terms of the chaotic turbulent condition of matter in such space. The changes also furnish fresh insight into problems of solar physics. However, interpretations of results of observations have been seriously handicapped by the paucity of data simultaneously obtained from observing stations widely distributed on the earth, and by non-

standardised experimental techniques followed at various places. It is fortunate that this state of affairs is now being remedied through the newly created Sub-Commission for Cosmic Ray Intensity Variations (SCRIV) and the programme in cosmic rays for the International Geophysical Year 1957–58. It is obviously important that worldwide studies should be continued without interruption through several solar cycles and that in future there should be several studies comparable with those made by Forbush at the Carnegie Institution of Washington.

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POLARIZATION OF FAST NUCLEONS[1]

By L. Wolfenstein

Department of Physics, Carnegie Institute of Technology, Pittsburgh, Pennsylvania

1. Introduction

Nuclear beams emerging from various types of accelerators are usually unpolarized; that is, the spin directions of the particles in the beam are randomly distributed. Recently an increasing number of experiments have been performed involving polarized beams in which the spins of the nucleons are preferentially oriented in a certain direction. The present article is primarily concerned with the theoretical formalism used in analyzing such experiments and with the results at high energies (nucleons between 100 and 400 Mev).

Specification of polarization.—The spin state of a spin-one-half particle is represented by a Pauli spinor $\binom{a_1}{a_2}$. For any such state there exists a direction $\mathbf{u}$ in which the spin is pointing, in the sense that if the z-axis had been chosen parallel to $\mathbf{u}$, the Pauli spinor would have $a_2=0$. If the unit vector $\mathbf{u}$ is defined by spherical coordinates θ_u, ϕ_u

$$a_1 = C \cos(\theta_u/2) e^{i\phi_u/2}$$
$$a_2 = C \sin(\theta_u/2) e^{i\phi_u/2} \qquad 1.$$

where $2|C|^2$ is the magnitude of the spinor.[2] A beam of spin-one-half particles is a mixture of many particles each of which may be considered as being in a spin state given by a Pauli spinor. If all the particles are in the same state the beam is said to be completely polarized and its polarization may be specified by a Pauli spinor; however, this is not generally the case, since the method of preparation usually does not completely polarize the beam.

We therefore must ask what is a proper procedure for specifying the polarization of a beam in general. For example, do we have to specify the spin states of each of the particles in the beam? What we require of our specification is that it completely determines the result of any experiments we may conceive. (It is assumed that the intensity and the momentum of the beam are also given.) For example, consider a Stern-Gerlach experiment with an inhomogeneous field along the z-direction. The difference between the fractions of particles ending up in the two beams would be given by

$$\frac{\sum_n \{|a_1^{(n)}|^2 - |a_2^{(n)}|^2\}}{\sum_n \{|a_1^{(n)}|^2 + |a_2^{(n)}|^2\}} = \overline{\langle \sigma_z \rangle} \qquad 2a.$$

where the sum over n is a sum over the states of all the particles in the original beam. $\langle \sigma_z \rangle$ is the expectation value of the Pauli operator for one of these states and the bar on top denotes an average over all the states. This

[1] The survey of literature pertaining to this review was completed in April, 1956.

[2] For later purposes it is convenient not always to normalize to unity.

experiment does not determine the results of Stern-Gerlach experiments with fields along the x- or y-directions (except in the special case of $\overline{\langle\sigma_z\rangle}=1$). The results of these additional experiments are given by

$$2\,\mathrm{Re}\sum_n a_1^{(n)*}a_2^{(n)}/\sum_n\{|a_1^{(n)}|^2+|a_1^{(n)}|^2\}=\overline{\langle\sigma_x\rangle} \qquad 2b.$$

$$2\,\mathrm{Im}\sum_n a_1^{(n)*}a_2^{(n)}/\sum_n\{|a_1^{(n)}|^2+|a_2^{(n)}|^2\}=\overline{\langle\sigma_y\rangle} \qquad 2c.$$

(It might be thought that one of these experiments is impossible because one of the axes is along the direction of motion, but the spin can be rotated with respect to the direction of motion by means of a magnetic field—(see Section 4)—before the Stern-Gerlach apparatus.) These three results determine a vector $\overline{\langle\boldsymbol{\sigma}\rangle}$ from which the result of an arbitrary Stern-Gerlach experiment can be found. Thus, the polarization of the beam may be specified by giving the direction of $\langle\boldsymbol{\sigma}\rangle$ and the amount of polarization P given by

$$P^2=\overline{\langle\boldsymbol{\sigma}\rangle}\cdot\overline{\langle\boldsymbol{\sigma}\rangle}=\overline{\langle\sigma_x\rangle}^2+\overline{\langle\sigma_y\rangle}^2+\overline{\langle\sigma_z\rangle}^2 \qquad 2d.$$

Since an arbitrary spin operator corresponding to any possible observation on the spin state may be represented as a linear combination of the unity operator, σ_x, σ_y and σ_z, its average value may be determined from Equation 2. This proves that $\overline{\langle\boldsymbol{\sigma}\rangle}$ is indeed the complete specification of the polarization which we desire. Two beams with the same value of $\overline{\langle\boldsymbol{\sigma}\rangle}$ cannot be distinguished even though they have been prepared in very different ways. For example, an unpolarized beam ($P=0$) may be considered as an equal mixture of any two completely and oppositely polarized beams, and any partially polarized beam ($1>|P|>0$) may be considered as a mixture containing a fraction $|P|$ of a beam completely polarized in the direction of $\overline{\langle\boldsymbol{\sigma}\rangle}$ and a fraction $(1-|P|)$ of an unpolarized beam.

This discussion is completely analogous to one that could be given for polarized light. The three Stern-Gerlach experiments are analogus to the three experiments—two to determine plane polarization and one to detect circular polarization—required to analyze polarized light. The three components of $\overline{\langle\boldsymbol{\sigma}\rangle}$ are direct analogues of the Stokes parameters (1, 2).

It may be noted in passing that the mean value of the spin vector is not sufficient for specifying the polarization of a particle of spin 1. In this case there are three beams in the Stern-Gerlach experiment, and so it is necessary to specify in addition the fraction of particles which are undeviated. This fraction may be expressed in terms of the mean value of an operator which transforms like an irreducible second rank tensor, and so in order to determine this fraction for all conceivable Stern-Gerlach experiments it is necessary to give five real numbers, making a total of eight parameters required to specify completely the polarization of a spin-one particle (3).

Double-scattering experiments.—The Stern-Gerlach experiment is not really a practical method for producing or analyzing polarization of fast nucleons because of the small magnitude of the magnetic moment. In addition, it may be shown (4) from the uncertainty principle that the usual semi-

classical discussion of the Stern-Gerlach experiment cannot be applied to a beam of charged particles such as protons. Another method, first proposed by Mott (5) for electrons, is to produce a polarized beam by scattering an unpolarized beam from a suitable target. If the scattering interaction couples the spin and orbital angular momenta, it is possible that the particles in the beam with spin up may be preferentially scattered to the left (right) while those with spin down may be preferentially scattered to the right (left). (It may be noted that classically left and right scattering from a repulsive central potential correspond to orbital angular momentum down and up, respectively.) A succession of two such scatterings, comprising a double-scattering experiment may be employed first to polarize the beam and then to analyze the polarization. The first successful experiment of this type was done by Shull (6) with electrons following the suggestion of Mott.

A simplified treatment of such an experiment is outlined in Table I.

TABLE I

INTENSITIES IN DOUBLE-SCATTERING EXPERIMENT
(SPIN-ZERO TARGET)

Beam	Spin Direction	Number of Particles
Incident	Up	N
	Down	N
Scattered once to left	Up	$Nf_1(1+P_1)$
	Down	$Nf_1(1-P_1)$
Scattered once to right	Up	$Nf_1(1-P_1)$
	Down	$Nf_1(1+P_1)$
Scattered twice to left (LL)	Up	$Nf_1f_2(1+P_1)(1+P_2)$
	Down	$Nf_1f_2(1-P_1)(1-P_2)$
Scattered first to left, then to right (LR)	Up	$Nf_1f_2(1+P_1)(1-P_2)$
	Down	$Nf_1f_2(1-P_1)(1+P_2)$

An unpolarized beam of $2N$ particles is scattered from the first target so that a fraction f_1 of all the particles reach target 2. However, it is assumed that for spin up particles scattered to the left[3] a fraction $f_1(1+P_1)$ reaches target 2, whereas for spin down particles scattered to the left the fraction is $f_1(1-P_1)$. Thus the beam striking target 2 has a polarization P_1 in the direction "up." The second scatterer is similarly characterized by f_2 and P_2, where P_2 represents the preference of target 2 for scattering spin up to the left. As a consequence it is seen that the beam which is twice scattered to

[3] In Figure 1 the scattering at target 1 is to the *left* and spin out of the page is spin *up*.

the left (LL) contains more particles than that scattered first to the left and then to the right (LR): the left-right asymmetry in the second scattering is given by

$$\epsilon = \frac{(\mathrm{LL}) - (\mathrm{LR})}{(\mathrm{LL}) + (\mathrm{LR})} = \frac{1 + P_1P_2 - (1 - P_1P_2)}{1 + P_1P_2 + (1 - P_1P_2)} = P_1P_2 \qquad 3.$$

Thus the parameter P determines both the polarizing power and the analyzing power of a target. In particular, if the two scatterings are identical, neglecting the loss of energy in the first scattering, we have $P_1 = P_2 = P$ and from Equation 3

$$P = \pm\epsilon^{1/2} \qquad 3a.$$

This gives us a method, the usual one employed at high energies, for determining the parameter P for a target and consequently the polarization of the once-scattered beam; however, it does not determine the sign of P. Note that the symbol P is used to represent both the amount of polarization of a beam and the polarizing power of a particular scattering.

While Equation 3 is correct the argument leading to it is incomplete. It has been tacitly assumed that particles starting with spin up end with spin up. This will be seen to be true in Section 3 for spin-zero targets, but it is not true in general for other targets. The general case will be discussed in Section 4, which also discusses triple-scattering experiments. The reader perhaps should also be warned that attempts to relate the sign of the polarization to the sign of the spin-orbit coupling by semi-classical arguments have only a 50 per cent chance of success.

Notation.—The quantities used to describe a single scattering are defined as follows: $\mathbf{k}$ unit vector in incident direction in lab; $\mathbf{k}'$ unit vector in outgoing direction in lab; $\mathbf{p}$ initial momentum in c.m. system; $\mathbf{p}'$ final momentum in c.m. system; Θ,ϕ spherical coordinates of $\mathbf{k}'$ with respect to k as polar axis; θ,ϕ spherical coordinates of $\mathbf{p}'$ with respect to p as polar axis; $\hbar\boldsymbol{\kappa}$ momentum transfer $= \mathbf{p}' - \mathbf{p}$; $\mathbf{K}$ unit vector in direction $\boldsymbol{\kappa}$; $\mathbf{P}$ unit vector in direction $(\mathbf{p}' + \mathbf{p})$; $\mathbf{n}$ unit vector normal to plane of scattering

$$\mathbf{n} = (\mathbf{k} \times \mathbf{k}')/\sin\Theta = (\mathbf{p} \times \mathbf{p}')/p^2 \sin\theta; \qquad 4a.$$

$$\mathbf{s}\ (\text{unit vector}) = \mathbf{n} \times \mathbf{k}'; \qquad 4b.$$

Subscripts 1, 2, and 3 will be used to indicate the first, second, and third scatterings in a sequence, but the subscript 2 may be omitted where this will cause no confusion. We may define the azimuthal angle for the second scattering by

$$\cos\phi_2 = \mathbf{n}_1 \cdot \mathbf{n}_2 \qquad \sin\phi_2 = \mathbf{n}_1 \times \mathbf{n}_2 \cdot \mathbf{k}_2 \qquad 4c.$$

2. Survey of Polarization Experiments

Suggestions for polarizing fast neutron beams by scattering were made by Schwinger (7, 8). One method depended on the large spin-orbit coupling

of the interaction of a neutron with a nucleus, which is evidenced by the large "fine structure" splitting of nuclear levels. In particular, the scattering of neutrons around 1 Mev from He^4 may be described in terms of a broad $J=3/2$ resonance corresponding to the ground state of He^5; the $J=1/2$ state appears to be at least 2 Mev higher in energy. Calculations have shown that the neutrons may be highly polarized as a result of the interference between the scattering from this split resonance and the potential scattering of the *s*-wave (9, 10). While this experiment has not as yet been performed with neutrons, the same method has been applied to protons by the Minnesota group (11, 12), who performed the first successful proton polarization experiment in 1951, producing and detecting the polarization by double-scattering from helium. Because of Coulomb effects, the corresponding $J=3/2$ resonance of Li^5, which is involved in this experiment, requires a proton energy between 2 and 3 Mev.

The fact that spin-orbit coupling is so prominent in nuclear interactions suggests that the products of many nuclear reactions may be polarized and that a variety of scattering experiments may be available for detection. This has been demonstrated by a number of successful experiments, including the following:

(*a*) The protons produced by the *d-d* reaction at a deuteron energy of 300 Kev have been shown to be polarized (13) by observing the left-right asymmetry when they are scattered from helium. Similarly the polarization of *d-d* neutrons has been observed using the resonance scattering from carbon or oxygen as an analyzer (14).

(*b*) The neutrons from the reaction Li^7 (p,n) Be^7 have been shown to have a polarization of about 50 per cent using scattering from oxygen as an analyzer (15). Polarized 400-Kev neutrons obtained in this way were scattered from a variety of targets from carbon to bismuth and in many cases a left-right asymmetry was observed (16).

General formulas for the polarization effects in nuclear reactions have been given by Blin-Stoyle (17a), Satchler (17b), and by Simon and Welton (18).

One of the suggested uses for fast polarized beams was a study of the spin dependence of the nucleon-nucleon interaction. As long as the interaction is a central one, even though there is a spin-spin interaction, the differential scattering cross section of a polarized beam will be the same as that of an unpolarized beam assuming the target is unpolarized. However, noncentral interactions which effectively couple spin and orbital angular momenta may result in a left-right asymmetry in the scattering of a polarized beam (19). This asymmetry cannot be large as long as the scattering is nearly all due to *s*-waves; therefore, polarized nucleon beams with energies well above 20 Mev are required.

The first successful attempt to polarize high-energy protons by scattering was made by Oxley and co-workers at Rochester in 1952 (20). Since then,

external proton beams with polarizations between 50 and 90 per cent and with intensities between 10^4 and 10^6 particles cm^{-2} sec^{-1} have been obtained from a number of cyclotrons at energies between 130 and 570 Mev (21 to 25). In a typical experimental arrangement (Fig. 1) the internal (unpolarized) beam is elastically scattered from target 1 of beryllium or carbon at an angle Θ_1 of between 10° and 20° and then is magnetically analyzed and collimated before striking target 2. In the usual double-scattering experiment the beam scattered at angle Θ_2 both to the left and to the right is counted with a

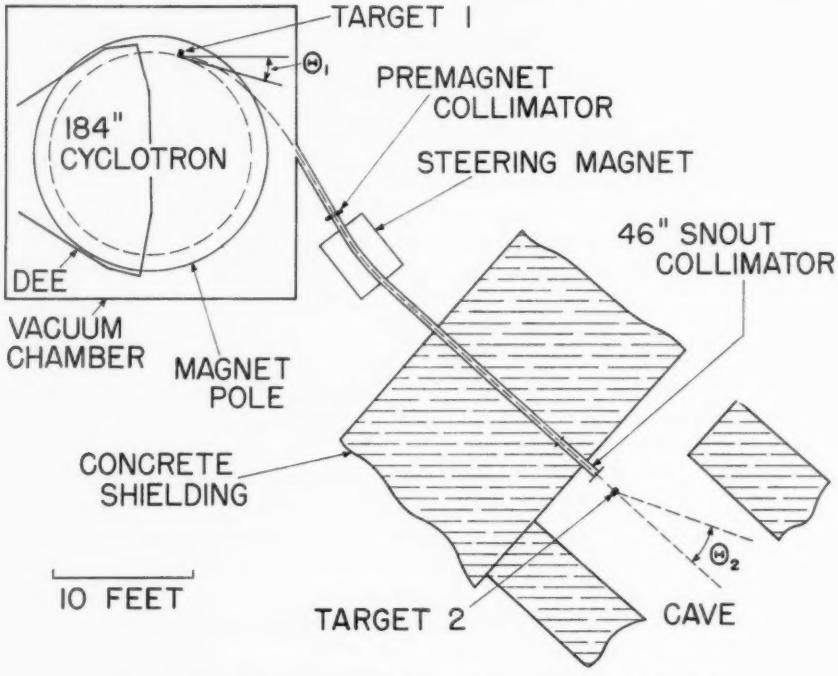

FIG. 1. Plan view of the Berkeley cyclotron showing orbit of the polarized beam (34).

counter telescope containing sufficient absorbing material so that only elastic or almost elastic scattering is counted. Since the interest is only in the left-right asymmetry it is not really necessary to determine the detection efficiency of the telescope. It is very important, however, to make certain that there are no intrinsic experimental asymmetries between left- and right-scattering. The results for scattering from complex nuclei and from hydrogen are discussed in Sections 3 and 5, respectively.

High energy neutron beams are usually obtained from the exchange scattering of protons with neutrons inside light nuclei. Viewing such a col-

lision inside the nucleus as a quasi-elastic n-p scattering, it would be hoped that the neutrons emerging at an angle to the incident proton direction are polarized by the noncentral n-p interaction. The first experiments to detect this polarization were done by Wouters (26) and demonstrated that the amount of polarization was not large. Neutron beams with a polarization of between 10 and 20 per cent have been obtained in this way with energies between 100 and 400 Mev (27 to 30) and have been used primarily to study n-p scattering (Section 5). The small magnitude of the polarization has the consequence that the asymmetry between left and right scattering is only a few per cent so that large errors may result from small misalignments of the experimental apparatus. The Harwell group (31) has recently overcome this difficulty by doing all the second scattering to one side first with the neutron spin "up" and then with the neutron spin "down." The neutron spin is reversed by sending it through a magnetic field about which the magnetic moment precesses.

It should be noted that the subject matter of the present article does not include polarized thermal neutrons or the use of stationary polarized nuclei as radioactive sources or as targets. Blin-Stoyle *et al.* cover these topics in their review article (32).

3. Scattering From a Spinless Target

We shall consider first the special case of the scattering of nucleons from a spin-zero nucleus such as helium or carbon. Actually experiments have indicated that the polarization of nucleons scattered from complex nuclei varies gradually as a function of atomic weight but seems to be independent of the spin of the nucleus (33). Also triple-scattering experiments from aluminum (34, 35) have shown that D (see Section 4) is approximately unity, the value it must have for spin-zero targets. Consequently, we follow the usual procedure and apply our discussion to scattering from any complex nucleus. The treatment is non-relativistic.

Use of the matrix scattering amplitude.—For this case the stationary state wave-function describing the elastic scattering may be written as a spinor with components

$$\psi_j^{(n)} = e^{ip_0 \cdot r/\hbar} a_j^{(n)} + \frac{e^{ipr/\hbar}}{r} \sum_l M_{jl}(\theta, \phi) a_l^{(n)} \qquad 5.$$

where $a_1^{(n)}$, $a_2^{(n)}$ define a particular spin state for the incident wave, and the 2×2 matrix $M(\theta, \phi)$, which relates the outgoing spinor to the incident one, takes the place of the usual scattering amplitude $f(\theta)$. The matrix M at any angle may be expanded in terms of Pauli matrices:

$$M = g1 + h_1\sigma_x + h_2\sigma_y + h_3\sigma_z = g + \mathbf{h} \cdot \boldsymbol{\sigma} \qquad 6.$$

where 1 is the unit matrix, which on the right and hereafter is not written explicitly. The x-, y-, and z-axes must be defined in terms of physical vec-

tors. For the scattering process, there are two of these: $\mathbf{p}$ and $\mathbf{p}'$. Since the only axial vector which can be constructed from these is $\mathbf{n}$, the most general form of M is

$$M = g(\theta) + h(\theta)\boldsymbol{\sigma}\cdot\mathbf{n} \tag{7.}$$

where g and h are arbitrary complex functions of energy and scattering angle θ. Terms like $\boldsymbol{\sigma}\cdot\mathbf{p}$ are ruled out because they change sign under space reflections.

If the initial spin direction $\boldsymbol{\mu}$ is defined by spherical coordinates θ_u, ϕ_u and the z-axis is chosen along $\mathbf{n}$, the spinor multiplying $(e^{ipr/\hbar}/r)$ in Equation 5 is from Equations 1 and 7.

$$\chi = \begin{pmatrix} C\,(g+h)\cos(\theta_u/2)e^{-i\phi_u/2} \\ C(g-h)\sin(\theta_u/2)e^{i\phi_u/2} \end{pmatrix} \tag{8.}$$

From Equation 8 we calculate

(a) the differential scattering cross section

$$I = \frac{\chi^+\chi}{2|C|^2} = |g|^2 + |h|^2 + 2\,\mathbf{n}\cdot\boldsymbol{\mu}\,\mathrm{Re}\,(g^*h) \tag{9.}$$

where $\mathbf{n}\cdot\boldsymbol{\mu}$ has been written in place of $\cos\theta_u$. Averaging over two opposite directions of $\boldsymbol{\mu}$, we obtain the cross section for an unpolarized incident beam

$$I_0 = |g|^2 + |h|^2 \tag{10a.}$$

The effect of the initial polarization is to add to the cross section a term I_p which may be written

$$I_p = I_0 P\,\mathbf{n}\cdot\boldsymbol{\mu} \tag{11a.}$$

$$P = 2\,\mathrm{Re}\,(g^*h)/(|g|^2 + |h|^2) \tag{10b.}$$

It is seen that for a given scattering angle θ, initial polarization along the direction $\mathbf{n}_1$ gives a $\cos\phi$ dependence to the cross section where ϕ is the usual azimuthal angle (Eq. 4c). Since I_0 and P determine the magnitudes of $(g+h)$ and $(g-h)$ we may write:

$$\begin{aligned} g + h &= [I_0(1+P)]^{1/2}\,e^{i(\alpha-\beta/2)} \\ g - h &= [I_0(1-P)]^{1/2}\,e^{i(\alpha+\beta/2)} \end{aligned} \tag{12.}$$

defining the phase factors α and β. Explicitly

$$\cos\beta = \frac{|g|^2 - |h|^2}{I_0(1-P^2)^{1/2}} \qquad \sin\beta = \frac{2\,\mathrm{Im}\,h^*g}{I_0(1-P^2)^{1/2}} \tag{10c.}$$

(b) the spin direction $\boldsymbol{\mu}_f$ of the scattered particle. The spherical coordinates θ_u', ϕ_u' defining $\boldsymbol{\mu}_f$ may be obtained by substituting Equation 12 into Equation 8 and comparing the result with Equation 1:

$$\begin{aligned} \phi_u' &= \phi_u + \beta \\ \cos\theta_u' &= (\cos\theta_u + P)/(1 + P\cos\theta_u) \end{aligned} \tag{13.}$$

In words, the spin of the particle is bent toward the z-axis (the normal $\mathbf{n}$) by an amount determined by P and the original z-component of spin, whereas

the direction of the projection of the spin in the scattering plane is rotated through the angle β. Averaging over two opposite directions of $\mathbf{y}$ we get for an unpolarized incident beam

$$\overline{\langle \boldsymbol{\sigma} \rangle}_t = P\mathbf{n} \qquad 11b.$$

The results of Table 1 follow directly from this discussion.

In order to determine β we must measure the rotation of the projection of the spin vector in the scattering plane. Such an experiment is illustrated in the second diagram in Figure 2. The scattering plane is chosen so as to contain the initial direction of polarization. Since a simple analyzer can only detect the component of the final polarization perpendicular to the outgoing direction $\mathbf{k}'$, the analyzer is chosen so as to detect the component of polarization along $\mathbf{s}$. If the incident beam is completely polarized this final component is called R (see Section 4) and for the present case

$$R = \sin\theta_u' \cos(\phi_u' - 90° - \Theta) = (1 - P^2)^{1/2} \cos(\beta - \Theta) \qquad 14.$$

where we have used Equation 13 with $\mathbf{k}$ as the x-axis, $\theta_u = 90°$, and $\phi_u = 90°$. (The other two components are $\langle \boldsymbol{\sigma} \rangle \cdot \mathbf{n} = P$ and $\langle \boldsymbol{\sigma} \rangle \cdot \mathbf{k}' \equiv R' = (1 - P^2)^{1/2}$ sin

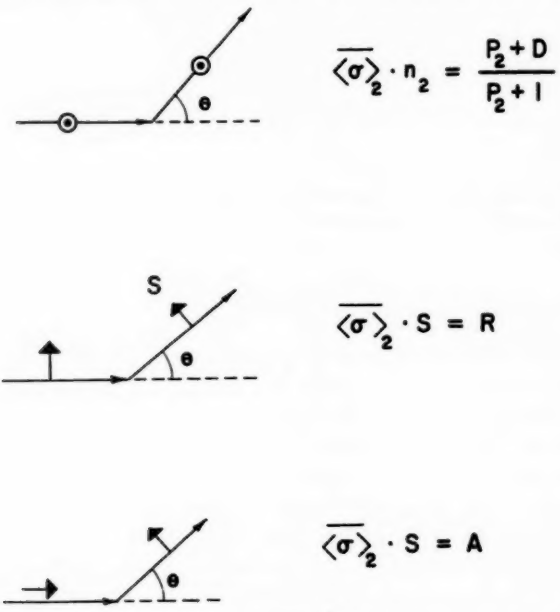
θ
$\overline{\langle \sigma \rangle}_2 \cdot n_2 = \frac{P_2 + D}{P_2 + 1}$
S
θ
$\overline{\langle \sigma \rangle}_2 \cdot S = R$
θ
$\overline{\langle \sigma \rangle}_2 \cdot S = A$

FIG. 2. Three triple scattering experiments. The direction of polarization is indicated by an arrow on the beam incident on the second scatterer (⊙ is an arrow out of the page). The arrow on the outgoing beam indicates the normal to the third scattering plane and thus the component of polarization to be measured. The equation for this component is given for the case in which the incident beam is completely polarized. The diagrams are in the laboratory reference frame.

$(\Theta - \beta)$.) The measurement of R thus involves three scatterings, the first polarizing the beam, the second the target of interest, and the third analyzing the final polarization. The third scattering plane is defined by being perpendicular to $\mathbf{s}$.

The single, double, and triple scattering experiments measure I_0, P and β and thus by Equation 12 determine at any scattering angle θ the complex amplitudes g and h except for the common phase factor α. This phase factor can only be determined by interference of the nuclear scattering with Coulomb scattering (except at $\theta = 0°$ where it may be determined from the optical theorem).

Scattering from a spin-orbit potential in Born approximation.—To calculate M we may assume that the incident nucleon interacts with the nucleus by means of a central potential augmented by a spin-orbit term

$$H_{\text{int}} = V(r) + W(r)\,\frac{\boldsymbol{\sigma}}{2}\cdot\frac{\mathbf{L}}{\hbar} \qquad 15a.$$

where $\mathbf{L}$ is the orbital angular momentum operator. In most theories it is convenient to express $W(r)$ in terms of a potential $Y(r)$

$$W(r) = -\left(\frac{\hbar}{mc}\right)^2 \frac{1}{r}\,\frac{dY}{dr} \qquad 15b.$$

so that

$$H_{\text{int}} = V(r) - \left(\frac{\hbar}{mc}\right)^2 \frac{1}{r}\,\frac{dY}{dr}\,\frac{\boldsymbol{\sigma}}{2}\cdot\frac{L}{\hbar} \qquad 15c.$$

Examples of such potentials are:

(*a*) The electromagnetic interaction of a moving nucleon with a nucleus of charge Ze in the nonrelativistic limit is given by Equation 15c with $V(r) = zZe^2/r$ and

$$Y(r) = -\frac{1}{2}(2\mu - z)\frac{Ze^2}{r} \qquad 15d.$$

where μ is the nucleon magnetic moment in units of the nuclear magneton and $z = 1$ for protons and 0 for neutrons.

(*b*) The interaction of a nucleon with a static potential $V(r)$ is augmented by a relativistic spin-orbit coupling due to the Thomas precession[4] given by $Y(r) = \frac{1}{2}V(r)$ (36, 37, 38). If $V(r)$ is the usual nucleon-nucleus interaction the magnitude of the spin-orbit coupling turns out to be much too small to explain experimental results, although recent theories employing an effective mass tend to reduce the discrepancy (39).

(*c*) The Mayer-Jensen shell model of the nucleus (40) requires a large spin-orbit coupling in the interaction of low energy nucleons with nuclei. A similar spin-orbit coupling term was proposed by Fermi (41) and others to

[4] It may be noted that the term $\frac{1}{2}zZe^2/r$ in Equation 15d may be identified as a result of the Thomas precession.

explain the high-energy polarization experiments. It has been argued by Fermi (42) that this spin-orbit coupling should be concentrated at the surface of the nucleus, since once a nucleon is surrounded by nuclear matter on all sides as far as it can "feel" it does not "know" its position with respect to the center of the nucleus and hence does not know the sign of **L**. This concentration may be achieved by using Equation 15b with $Y(r)$ directly proportional to $V(r)$. Various attempts have been made to relate this spin-orbit coupling to the noncentral part of the nucleon-nucleon interaction (43 to 47).

We now calculate in the Born approximation the contribution M_{LS} to M from the second term in Equation 15c for the scattering from momentum $\mathbf{p}$ to $\mathbf{p}'$.

$$M_{LS} = \frac{m}{2\pi\hbar^2}\left(\frac{\hbar}{mc}\right)^2 \int e^{-i\mathbf{p}'\cdot\mathbf{r}/\hbar}\,\frac{1}{r}\,\frac{dY}{dr}\,\frac{\boldsymbol{\sigma}}{2}\cdot\mathbf{r}\times\frac{\nabla}{i}\,e^{i\mathbf{p}\cdot\mathbf{r}/\hbar}\,d\tau$$

$$= -\frac{m}{2\pi\hbar^2}\left(\frac{\hbar}{mc}\right)^2 \frac{\boldsymbol{\sigma}}{2}\cdot\frac{\mathbf{p}}{\hbar}\times\int e^{-i\boldsymbol{\kappa}\cdot\mathbf{r}}\nabla Y(r)\,d\tau.$$

Integrating by parts:

$$M_{LS} = \frac{m}{2\pi\hbar^2}\left(\frac{\hbar}{mc}\right)^2 \frac{\boldsymbol{\sigma}}{2}\cdot\frac{\mathbf{p}}{\hbar}\times\int\nabla(e^{-i\boldsymbol{\kappa}\cdot\mathbf{r}})Y(r)\,d\tau$$

$$= \frac{i}{2}\left(\frac{p}{mc}\right)^2 \sin\theta\; y(k)\;\boldsymbol{\sigma}\cdot\mathbf{n} \qquad 16.$$

where we have noted that

$$\mathbf{p}\times\hbar\boldsymbol{\kappa} = \mathbf{p}\times\mathbf{p}' = p^2\sin\theta\,\mathbf{n}$$

and the notation for a Fourier transform has been introduced:

$$x(\kappa) = -\frac{m}{2\pi\hbar^2}\int e^{-i\boldsymbol{\kappa}\cdot\mathbf{r}}\,X(r)\,d\tau$$

Here p and κ are the magnitudes of $\mathbf{p}$ and $\boldsymbol{\kappa}$, respectively. The final Born approximation result then is

$$g(\theta) = v(\kappa) \qquad 17a.$$

$$h(\theta) = i\eta^2\sin\theta\; y(\kappa)/2 \qquad 17b.$$

$$\hbar\kappa = 2p\sin(\theta/2) \quad \eta = p/mc$$

A physical picture of this Born approximation calculation has been given by Fermi (42).

The interpretation of the scattering of nucleons from nuclei in terms of the optical model (48, 49, 50) requires the use of a complex potential $V(r)$, in which the imaginary part accounts for absorption:

$$V(r) = V_0(r)(1+i\epsilon) \qquad 18.$$

We shall follow the usual assumption that the spin-dependent part of the potential is real and given by Equation 15b with

$$V(r) = \gamma V_0(r) \qquad 19.$$

although the use of a complex $Y(r)$ has recently been suggested both on theoretical grounds (47) and as a means of fitting experimental results (51). Using Equations 18 and 19 for the potential we obtain (41, 52, 53) from Equations 10 and 17

$$I_0 = v_0^2(\kappa)[1 + \epsilon^2 + (\gamma^2\eta^4 \sin^2\theta)/4$$

$$P = \frac{\eta^2\gamma\epsilon \sin\theta}{1 + \epsilon^2 + \gamma^2\eta^4 \sin^2\theta/4} \qquad 20.$$

$$\sin\beta = -(1/\epsilon)\{P/(1 - P^2)^{\frac{1}{2}}\}$$

It should be noted that the polarization P vanishes if the potentials $V(r)$ and $Y(r)$ are both real, (that is $\epsilon = 0$); in this case the scattering amplitudes for spin "up" and spin "down" differ in phase but not in magnitude, since h is 90° out of phase with g. The vanishing of the polarization in the Born approximation for a Hermitean Hamiltonian may be proven in general [cf. footnote 13 in Ref. (18a)]. In this case, the spin-orbit coupling still displays itself in the triple-scattering as may be seen by substituting the equation for (P/ϵ) into the equation for $\sin\beta$.

More exact calculations (45, 54 to 61), most of them based on the WKB method, have shown that the Born approximation results may be completely incorrect near diffraction minima but can be used as a rough guide elsewhere.

Comparison of theory and experiment.—We wish to compare the results of the complex potential model with spin-orbit coupling to the experimental results around 300 Mev (34). In the Born approximation the polarization as a function of angle is seen (Equation 20) to be independent of $V_0(r)$; it reaches a maximum value of

$$P_{max} = \epsilon/(1 + \epsilon^2)^{1/2} \qquad 21a.$$

at an angle θ_{max} given by

$$\sin\theta_{max} = 2(1 + \epsilon^2)^{1/2}/\gamma\eta \qquad 21b.$$

Applying Equation 21 to the data for 313-Mev protons on carbon gives ϵ about 1.0 and γ about 16; this value of γ is more than thirty times the value of one-half given by the Thomas precession. The Born approximation result (41) for $\gamma = 15$ and $\epsilon = 0.6$ is compared with the experimental data in Figure 3.

In an exact calculation the polarization is dependent on the potential $V_0(r)$. If the same values $\gamma = 15$ and $\epsilon = 0.6$ are used and a square well of radius 3.2×10^{-13} cm. and depth 27 Mev assumed for $V_0(r)$, an exact calculation yields the polarization shown by the solid curve in Figure 3 (54). For comparison with the data the experimental angular resolution has been folded into the theoretical curve. The exact results are characterized by a large fluctuation of the polarization in the region of the expected diffraction minimum. It is easy to see why one might expect such a fluctuation. Consider the scattering of a completely polarized beam: the particles scattered to the left see effectively a different potential than those scattered to the right and as a result the diffraction minimum on the left occurs at a some-

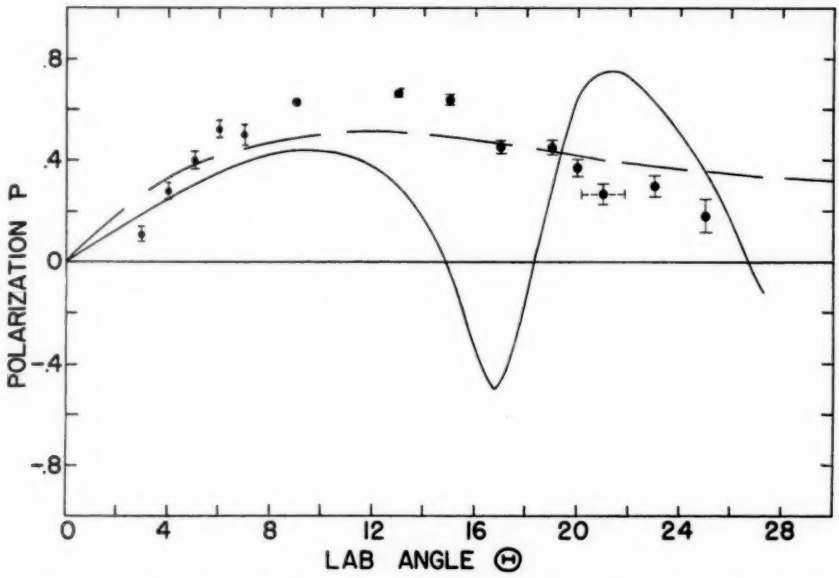

FIG. 3. Polarization P as a function of laboratory angle for scattering of 313 Mev protons from carbon (34). Solid curve is an exact calculation (54) (with angular resolution folded in) using a square-well potential; dashed curve is the Born approximation result for the same potential (41).

what smaller scattering angle than on the right. When the diffraction minimum occurs on the left, nearly all the particles scatter to the right and we have a large negative value of P; at a slightly larger angle the situation is reversed and we have a large positive value of P. This is illustrated in Figure 4, which shows the left and right scattering cross sections for 76 per cent polarized protons according to this exact calculation. It is only for the Born approximation calculation with Equation 19 that the left- and right-scattering cross sections display identical diffraction patterns.

In contrast to these exact theoretical results the experiments on carbon do not show a diffraction minimum in the cross section nor a large dip in the polarization. Much better fits to the data have been obtained (45, 57, 61) by replacing the square well used for $V_0(r)$ by a more gentle shape as had already been suggested by Woods & Saxon (49) in order to fit cross section data around 20 Mev. The fluctuation in the polarization remains with reduced magnitude in these calculations, and careful experiments on aluminum at 313 Mev (34) have shown that such fluctuations do indeed occur. Sternheimer (57) fits the data on aluminum using a rounded-edge Woods-Saxon potential for $V_0(r)$ and Equations 18 and 19 with $\epsilon=1$ and γ about 12 (Fig. 5). It should be noted that the theoretical differential cross section and triple-scattering results using the same potential do not appear to fit the

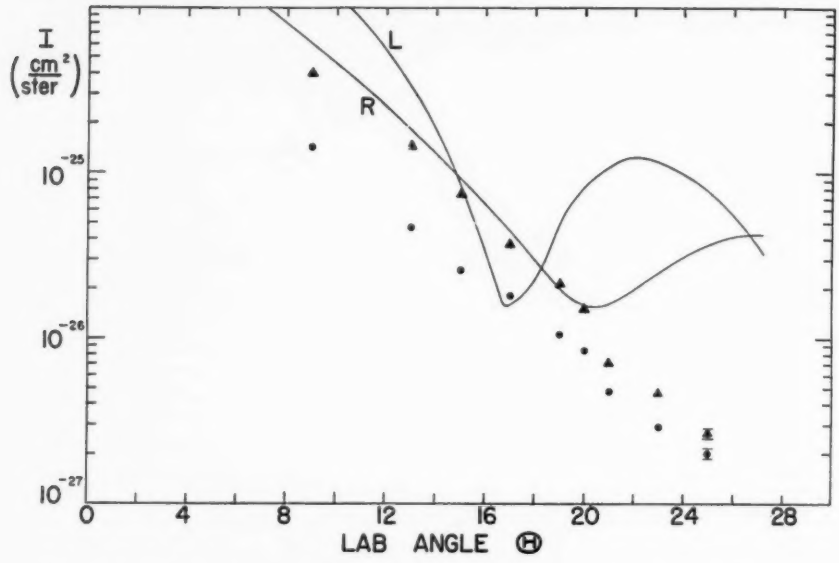

FIG. 4. Left (triangles) and right (circles) differential cross-sections of 76 per cent polarized 313 Mev protons scattered from carbon (35). Solid curves are an exact calculation (with angular resolution folded in) using the same potential as in Fig. 3.

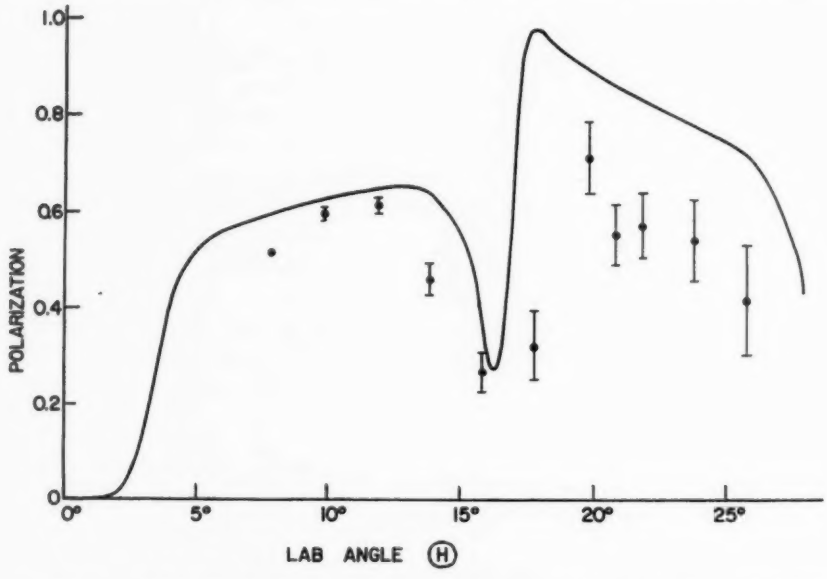

FIG. 5. Polarization P of 300 Mev protons scattered from aluminum (34). The solid curve represents the theoretical fit made by Sternheimer (57). Angular resolution of about 1° is not considered in the theoretical curve.

data so well. Actually considerable caution is necessary in comparing experimental and theoretical results near the theoretical diffraction minimum and also at larger angles, even if the experimental angular resolution is folded in (which was not done in Fig. 5). The reason is that most experiments fail to resolve inelastic scattering (to the first few excited states) from elastic scattering; this inelastic scattering may have a maximum cross section in the region of the elastic cross section minimum (62, 63). Thus the observations at these angles may really be primarily observations on inelastic scattering. In recent experiments on carbon with 220 Mev polarized protons (64) inelastic scattering to the first excited state is resolved and found to be considerably larger than the elastic scattering for angles above 25°. The polarization $P(\theta)$ for pure elastic scattering is shown to have large fluctuations, actually going slightly negative between 25° and 30° and then rising to a value close to 1 around 35°, a behavior characteristic of most of the theoretical calculations.

Triple scattering experiments have been done at 300 Mev on carbon and aluminum to determine the parameter β (34). Theoretical calculations (57) tend to give values of β fluctuating rapidly with angle. In contrast, the experimental results give an almost constant value of β between 8° and 22° for aluminum; however, significant fluctuations could have been missed because of the angular resolution.

Energy dependence of the polarization.—Similar results for the polarization in scattering from complex nuclei have been obtained at energies from 130 Mev to 420 Mev. In Figure 6, the maximum polarization P_{max} and the corresponding scattering angle Θ_{max} are plotted as a function of energy for scattering from carbon. The following comments should be made about the figure: (*a*) The black circles represent experiments in which the polarization was measured at only one or two angles, not necessarily at Θ_{max}; and so these give only a lower limit to P_{max}. (*b*) The determination of the absolute value of P requires a knowledge of the polarization of the beam, which may be uncertain because of the difficulty of ascertaining the scattering angle of the first scattering which takes place inside the cyclotron (Fig. 1). An estimate of this error has been compounded in the errors indicated in Figure 6. Since the polarization is retained when a high-energy beam is degraded to a lower energy (9) it is possible for one laboratory to check its determination of beam polarization with that of another laboratory; a number of such experiments (65, 66, 67) indicate good agreement so that the comparison of results from different laboratories should be all right. (*c*) Values of P_{max} could be too low because of the finite angular resolution (usually $\pm 1°$) of the experiments or because of inelastic events included. (*d*) The errors in Θ_{max} are primarily due to the flatness of the maximum of $P(\Theta)$, which makes it hard to pick out Θ_{max}.

The following features of Figure 6 appear of interest: (*a*) P_{max} is above 0.8 and fairly constant between 130 Mev and 240 Mev, while Θ_{max} varies approximately inversely as the energy. This is consistent with the Born approximation calculations (Equation 21) with fixed values of the optical

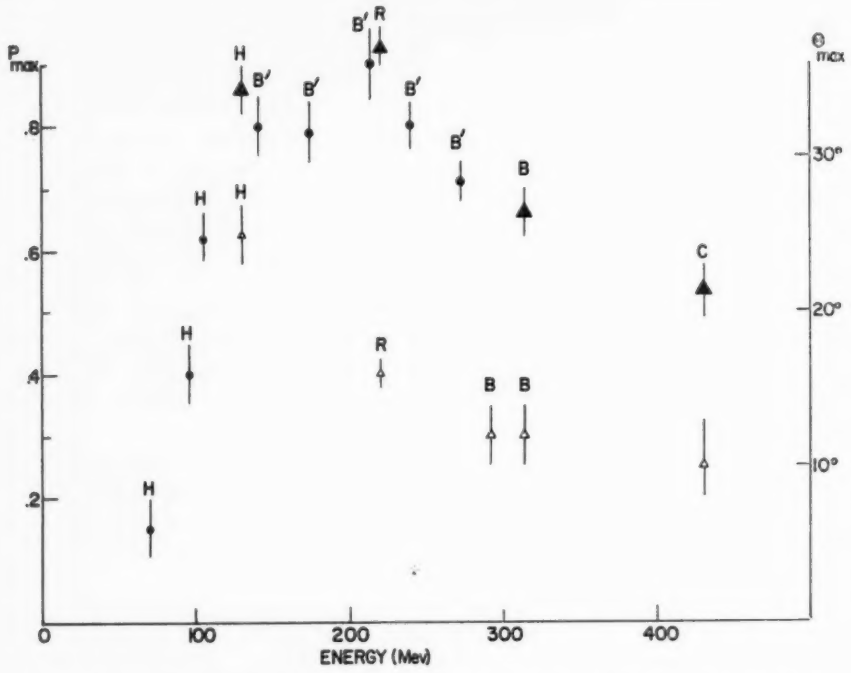

FIG. 6. Maximum polarization P_{max} and the scattering angle Θ_{max} for maximum polarization in scattering from carbon. Black triangles—P_{max}. Black circles—Lower limits to P_{max}. White triangles—Θ_{max}. C—Carnegie Tech (65). B—Berkeley (34). B′—Berkeley (66). R—Rochester (64). H—Harwell (33). (Recent results raise the Harwell P_{max} at 135 Mev to $.96_t$ .03 and also the point at 70 Mev to .3.)

parameters ϵ and γ. (*b*) From somewhere around 250 Mev, P_{max} starts to decrease gradually at least up to 420 Mev, while Θ_{max} continues to decrease slowly. This behavior could be explained in the Born approximation by a decrease of both ϵ and γ by a factor of about two. No more exact study of this variation with energy seems to have been made. However, the work of Watson (47) suggests that an imaginary part of $W(r)$ may play an important role in this energy region. (*c*) Below 130 Mev the polarization appears to fall off rapidly; indeed attempts to produce a polarized beam by scattering at 80 Mev (68) proved unsuccessful. It should be emphasized, however, that the values shown below 130 Mev are all at 30° and it would seem likely that higher polarizations could be obtained by scattering at larger angles. Some indirect evidence that this indeed is the case comes from the analysis (3) of the polarization of deuterons between 100 Mev and 200 Mev scattered from carbon (69). Sternheimer (58) has shown that the decrease in polarization below 130 Mev may be explained quantitatively by the decrease in the optical parameter ϵ as the energy is decreased; such a decrease had al-

ready been suggested (70, 71) to fit neutron scattering measurements. However, even with this decreased value of ϵ large polarizations (greater than 50 per cent) may be possible at certain angles, as has been shown by calculations by Culler *et al.*, for 14-Mev scattering (72). Furthermore, the polarization of the order of 20 per cent observed (16) in the scattering of 0.5 Mev neutrons can be explained using the very small value of ϵ from the cloudy-crystal ball model (50).

Dependence of polarization on target.—Polarized protons have been scattered from a variety of targets from helium through tantalum at 300 Mev (34) and from beryllium through bismuth at 130 Mev (33). The general trend of the results as the atomic number is increased from carbon to iron appears to be a gradual lowering of the value of P_{max} at the first polarization maximum together with the appearance of a second maximum at somewhat larger angles. The lowering of the value of P_{max} has been attributed to Coulomb effects (45), while the fact that the magnitude changes only very slowly as a function of nuclear radius has been cited (60) as evidence for the concentration of the spin-orbit interaction at the nuclear surface.

Sign of the polarization.—If it is assumed that γ is positive in accordance with the shell model it follows that P is positive, where the sign convention is given by Equations 11 and 4a; this means that nucleons with spin up scatter preferentially to the left. This result may be seen to follow if the Born approximation is used to calculate $h(\theta)$ using a real spin-orbit coupling potential: then from Equations 17b, 10b, and 19

$$I_0 P = \eta^2 \gamma \sin\theta \, v_0(\kappa) \operatorname{Im} g(\theta)$$

and at small angles Im $g(\theta)$ is positive since Im $g(0)$ is given by the optical theorem; thus the sign of P at small angles is the same as the sign of γ. The double-scattering experiments discussed so far determine only the product P_1P_2 and thus are noncommital on the sign of P. In order to determine the sign of P, polarized protons have been slowed down to energies below 10 Mev and scattered from helium (see Section 1); the sign of the parameter P is known for this case from a detailed phase shift analysis (11). The result of these experiments has been to show that the polarization is indeed positive (73, 74). It is interesting to note that the same sign of polarization is observed for small-angle p-p and n-p scattering (Section 5).

Electromagnetic scattering.—In addition to the specific nuclear interactions we must consider the scattering due to the Coulomb field of the nucleus. This should be particularly important at small angles or near diffraction minima. The scattering matrix M_c due to the Coulomb field includes a spin-dependent part due to the interaction of the nucleon magnetic moment (cf. Equation 15d) and is given for an infinitely heavy point nucleus with low atomic number Z by Equation 7 with

$$g_c(\theta) = -\frac{1}{2}(n/\eta)(\hbar/mc)\left\{z \csc^2\left(\frac{\theta}{2}\right) - \frac{\eta^2}{E}\mu_1\right\} \exp\left[2iz\left\{n \ln \csc\left(\frac{\theta}{2}\right) + \eta_0\right\}\right] \quad 22a.$$

$$h_c(\theta) = \frac{i}{2}\left(\frac{Ze^2}{\hbar c}\right)\left(\frac{\hbar}{mc}\right)\mu_1 \cot\left(\frac{\theta}{2}\right) \exp\left[2iz\left\{n \ln \csc\left(\frac{\theta}{2}\right) + \eta_0\right\}\right] \qquad 22b.$$

where

$$n = Ze^2/\hbar v = (Ze^2/\hbar c)(E/\eta)$$
$$E = \eta^2 + 1 = (\text{total energy})/mc^2$$
$$\mu_1 = \mu - zE/(E+1)$$
$$\eta_0 = \arg \Gamma(1 + in)$$

Equation 22 may be derived without the exponential phase factor by applying the Born approximation to the Dirac equation including a Pauli moment. It has been derived with the phase factor by Garren (75) as the first term in an expansion in powers of n. A similar result for small angles and low velocities has been found in the WKB approximation by Heckrotte (51).

The simplest application is to the polarization of neutrons ($z=0$) scattered at such small angles that the magnetic moment scattering $h_c(\theta)$ has the same order of magnitude as the nuclear scattering. In this case the Born approximation result should be valid even for large Z (8). From Equations 22b and 10b we obtain

$$I_0 P = (Ze^2/\hbar c)\mu\eta \cot(\theta/2) \operatorname{Im}(\hbar g_N(\theta)/p) \qquad 23.$$

where $g_N(\theta)$, the nuclear scattering, is assumed to be spin-independent at these small angles. The factor Im $[\hbar g_N(\theta)/p]$ can be approximated at these small angles by its value at 0° which is equal to the transmission cross section divided by 4π. It is thus possible to determine all the factors in Equation 23 and so make a theoretical calibration of an analyzer for polarized neutrons. This method has been used by Voss & Wilson (30) who have determined the sign and magnitude of the polarization of their 100-Mev neutron beam by scattering from uranium at angles between $\frac{1}{3}$° and 1°.

Coulomb effects for the case of proton scattering have been included in a number of calculations (including those of Sternheimer illustrated in Figure 5) and influence markedly the fluctuation in $P(\theta)$ near the expected diffraction minimum. However the magnetic moment effect has been ignored in all the calculations [which follow the method of Gatha & Riddell (76)] except for the analysis by Heckrotte (51) of the polarization of 300-Mev protons scattered from carbon at small angles (34). In this analysis $I_0P(\theta)$ is a sum of the purely nuclear term and a positive term similar to Equation 23 due to the interference of $h_c(\theta)$ and $g_n(\theta)$; in order to fit the shape of the experimental $P(\theta)$ curve, between 2° and 6° it is concluded that these two terms must have the same sign. This result provides some additional confirmation for the positive sign of the polarization as a result of the purely nuclear scattering.

4. General Formalism

In Section 3 we considered a well defined initial spin state yielding a well defined final spin state and we averaged over initial spins or summed over final spins where necessary. When the target also has a spin this procedure

becomes too awkward and a more general formalism which emphasizes operators rather than wave functions becomes extremely desirable.

Density matrix formalism.—(77, 78). We consider the system of two particles with spins s and s_t and discuss first the specification of the polarization, generalizing the discussion of Section 1. An arbitrary spin state (n) is a linear combination of the $(2s+1)\ (2s_t+1)$ basic states of the composite system and may be represented by a vector $a_i^{(n)}$ where i runs from 1 to $(2s+1)\ (2s_t+1)$. For example, if $s=s_t=\frac{1}{2}$, the four basic states could be the three triplet and one singlet states. An arbitary operator may be written as a linear combination of Hermitean base matrices, S^μ, where μ runs from 1 to $(2s+1)^2\ (2s_t+1)^2$. We require for these the orthogonality relations

$$\mathrm{Tr}\ (S^\mu S^\nu) = (2s+1)(2s_t+1)\ \delta_{\mu\nu} \tag{24.}$$

For example, for a spin-one-half particle incident on a spin-one-half target, the sixteen base matrices could be $1 1_t$, $\sigma_\alpha 1_t$, $1 \sigma_{t\alpha}$, $\sigma_\alpha \sigma_{t\beta}$, where the first matrix operates on the incident-particle spinor, the second matrix (with subscript t) operates on the target spinor, and α and β may take the values x, y, and z. The expectation value $\langle S^\mu \rangle_n$ of one of these matrices for the state (n) is given by

$$\sum_j |\, a_j^{(n)} \,|^2 \langle S^\mu \rangle_n = (a_1^{(n)*}\, a_2^{(n)*} \cdots) \begin{pmatrix} S_{11}^\mu & S_{12}^\mu & \cdots \\ S_{21}^\mu & \cdots & \\ \cdots & & \end{pmatrix} \begin{pmatrix} a_1^{(n)} \\ a_2^{(n)} \\ \cdot \end{pmatrix}$$

$$= \sum_k \sum_j a_j^{(n)}\, a_k^{(n)*}\, S_{kj}^\mu \tag{25.}$$

where $\sum_j |\, a_j^{(n)} \,|^2$ represents the relative probability or intensity of the state (n).

Any vector $a_i^{(n)}$ defines a completely polarized state; that is, one in which each of the particles is in a well defined spin state. In general neither of the particles will be completely polarized, so we must average over all the states (n) of the composite system which are incoherently mixed together, weighting each with its relative probability $\sum_j |\, a_j^{(n)} \,|^2$. In particular, from Equation 25 we obtain such an average for the expectation value of S^μ:

$$\overline{\langle S^\mu \rangle} = \frac{\sum_n \sum_k \sum_j a_j^{(n)}\, a_k^{(n)*} S_{kj}^\mu}{\sum_n \sum_j |\, a_j^{(n)} \,|^2} \tag{26.}$$

The averages $\overline{\langle S^\mu \rangle}$ for all values of μ may be used to specify the polarization of the composite system (cf. the discussion below Equation 2d). Defining the density matrix[5] (79)

$$\rho_{jk} = \sum_n a_j^{(n)}\, a_k^{(n)*} \tag{27.}$$

[5] It should be noted that the sum over n is over all the states that have been incoherently mixed together by the method of preparation so that the sum may include nonorthogonal states and indeed the same state may enter several times in the sum. For a beam of particles the sum may be considered as a sum over the states of the individual particles in the beam.

we have

$$\langle \overline{S^\mu} \rangle = \mathrm{Tr}\,(\rho S^\mu)/\mathrm{Tr}\,(\rho) \qquad 28.$$

From Equations 24 and 28 we obtain an expansion of ρ in terms of the S^μ

$$\rho = \mathrm{Tr}\,(\rho) \sum_\mu \langle \overline{S^\mu} \rangle\, S^\mu/[(2s+1)(2s_t+1)] \qquad 29.$$

The density matrix is therefore a compact method of specifying the polarization. One of the S^μ is always taken as the unit matrix whose average value is unity so that the polarization is specified by $[(2s+1)^2(2s_t+1)^2-1]$ real numbers. This should be contrasted with the $[2(2s+1)(2s_t+1)-2]$ numbers required to specify a completely polarized state (not including its normalization or overall phase). The factor $\mathrm{TR}(\rho)$ defines the normalization of the density matrix and is conveniently taken proportional to the intensity.

For a nucleon and a spinless target, $s=\frac{1}{2}$, $s_t=0$, the S^μ are 1, σ_x, σ_y, σ_z, and the density matrix is by Equation 29

$$\rho = \tfrac{1}{2}\,\mathrm{Tr}\,(\rho) \begin{bmatrix} 1+\langle \overline{\sigma_z} \rangle & \langle \overline{\sigma_x} \rangle - i\,\langle \overline{\sigma_y} \rangle \\ \langle \overline{\sigma_x} \rangle + i\,\langle \overline{\sigma_y} \rangle & 1-\langle \overline{\sigma} \rangle_z \end{bmatrix}$$

the density matrix for an unpolarized system is a multiple of the unit matrix, while a necessary and sufficient condition for a completely polarized system is $\mathrm{Tr}(\rho^2)=(\mathrm{Tr}\rho)^2$.

The wave function describing elastic scattering corresponding to any initial spin state $a_j^{(n)}$ can be expressed as before by Equation 5. When the initial colliding beams contain an incoherent mixture of states (n), the final outgoing beams for some scattering direction (θ, ϕ) will also contain a mixture of states, which can be described by a density matrix ρ_f obtained from Equations 5 and 27:

$$\rho_f)_{jk} = \sum_n \Big(\sum_l M_{jl}\, a_l^{(n)}\Big)\Big(\sum_r M_{kr}\, a_r^{(n)}\Big)^*$$

$$= \sum_l \sum_r M_{jl}\, \rho_i)_{lr}\, M_{kr}^*$$

or in matrix notation

$$\rho_f = M\rho_i M\dagger \qquad 30.$$

where ρ_i is the initial density matrix. Substituting Equation 30 in Equation 28, and using Equation 29 for ρ_i we find

$$I\langle \overline{S^\mu} \rangle_f = \sum_\nu \langle \overline{S^\nu} \rangle_i\, \mathrm{Tr}\,(MS^\nu M\dagger S^\mu)/[(2s+1)(2s_t+1)] \qquad 31.$$

where

$$I = \mathrm{Tr}\,(\rho_f)/\mathrm{Tr}\,(\rho_i) \qquad 31a.$$

is the differential scattering cross section.

Equation 31 may be extended to the case of a reaction process by letting M be a matrix (in general rectangular) which transforms from the spin space of the initial channel to the spin space of the final channel. [For a special case, see Ref. (80).]

Double-scattering experiment.—This experiment may be described in terms of two special cases of Equation 31:

(*a*) In the second scattering (analyzer) we are interested in the differential scattering cross section I for the case of a polarized nucleon beam with $\langle \vec{\sigma} \rangle_i = P_i \mathbf{y}$ incident on an unpolarized target of spin s_t:

$$I = \{\mathrm{Tr}\,(MM\dagger) + P_i \mathbf{u} \cdot \mathrm{Tr}\,(M\boldsymbol{\sigma}M\dagger)\}/2(2s_t + 1)$$
$$= \mathrm{I}_0 + P_i I_p \qquad 32.$$

The first term I_0, which comes from letting S^ν on the right-hand side of Equation 31 equal the unit matrix, is the differential cross section of an unpolarized beam. The second term $P_i I_p$, which comes from letting S^ν equal the three components of $(\boldsymbol{\sigma} 1_t)$, is the contribution to the cross section of the initial polarization of magnitude P_i. All other $\langle \overline{S^\nu} \rangle_i$ are zero because the target is unpolarized. Note that the results for the spinless target case (Equations 10a, 11a, and 10b) may be obtained by substituting Equation 7 into Equation 32.

(*b*) In the first scattering (polarizer) an unpolarized beam hits an unpolarized target. In this case all $\langle \overline{S^\nu} \rangle_i$ equal zero except the unit matrix, and the polarization of the scattered nucleon beam is

$$I_0 \langle \vec{\sigma} \rangle_f = \mathrm{Tr}\,(MM\dagger\boldsymbol{\sigma})/2(2s_t + 1) \qquad 33.$$

We now state three theorems governing these quantities:

Theorem 1. (9) (*a*) The contribution I_p of the initial polarization (in direction $\mathbf{y}$) to the differential cross section always has an azimuthal dependence proportional to $\cos \phi$ $(= \mathbf{n} \cdot \mathbf{y})$. It follows that polarization parallel to the direction of motion cannot be detected by a single scattering alone. (*b*) The polarization $\langle \vec{\sigma} \rangle_f$ (Equation 33) produced by the polarizer is along the normal $\mathbf{n}$ to the scattering plane.

Theorem 2. (77, 78). The quantities I_p and $\langle \vec{\sigma} \rangle_f$, which describe the analyzing power and polarizing power, respectively, may be expressed in terms of a single scattering parameter $P(\theta)$ by means of Equations 11a and 11b (derived in Section 3 for the spinless-target case). Comparing these with Equations 32 and 33 we may express this theorem

$$\mathrm{Tr}\,M\boldsymbol{\sigma}M\dagger = \mathrm{Tr}\,MM\dagger\boldsymbol{\sigma} = 2(2s_t + 1) I_0 P \mathbf{n} \qquad 34.$$

The first equality is not at all obvious since M, $M\dagger$, and $\boldsymbol{\sigma}$ do not commute with one another. It follows that for a double-scattering experiment the differential cross section for the second-scattering

$$I_2 = I_{02}(1 + P_1 P_2 \cos \phi_2) \qquad 35.$$

where the subscripts 1 and 2 describe the scattering angle, energy, etc., of the first and second scatterings respectively. Equation 3 for the asymmetry e follows immediately.

Theorem 3. (9) If $L_{\max}$ is the maximum orbital angular momentum that need be considered the angular distribution of the polarization must be of the form

$$I_0 P = \sum_{n=0}^{2L_{\max}-1} a_n \cos^n \theta \sin \theta \qquad 36.$$

The proof of these theorems depends on the invariance of M under space rotations and reflections and time-inversion. We may write the matrix M in the form of Equation 6, where g and $\mathbf{h}$ are now operators (depending on $\mathbf{p}$ and $\mathbf{p}'$) in the spin-space of the target nucleus. The invariance conditions tells us that g transforms like a scalar and does not change sign under time-inversion, whereas $\mathbf{h}$ must transform like an axial vector and change sign under time-inversion in order that $\mathbf{h}\cdot\boldsymbol{\sigma}$ be invariant. Using Equation 6 we obtain

$$\begin{aligned} \mathrm{Tr}\, M\sigma M\dagger &= 2\,\mathrm{Tr}'\,(\mathbf{h}g\dagger + g\mathbf{h}\dagger - i\mathbf{h}\times\mathbf{h}\dagger) \\ \mathrm{Tr}\, MM\dagger\sigma &= 2\,\mathrm{Tr}'\,(\mathbf{h}\dagger g + g\dagger\mathbf{h} - i\mathbf{h}\dagger\times\mathbf{h}) \end{aligned} \qquad 37.$$

Here Tr' stands for the trace in the target spin space, since we have already taken the trace in the nucleon spin-space. After we take the indicated traces in Equation 37 we are left with functions of $\mathbf{p}$ and $\mathbf{p}'$. We note that the last terms of each of these traces must transform like an axial vector and must not change sign under time reversal (since they involve $\mathbf{h}$ twice). Since the only axial vector we can form, $(\mathbf{p}\times\mathbf{p}')$, does change sign under time reversal, the last term in each of the traces must vanish. The remaining terms which are linear in $\mathbf{h}$ must be proportional to $(\mathbf{p}\times\mathbf{p}')$ $(=\mathbf{n}p^2\sin\theta)$, whence Theorem 1 follows from Equations 32 and 33. The $\sin\theta$ dependence in Equation 36 also follows from this argument.[6] This means that if we expand $\mathbf{h}$ in terms of components in Equation 37, only the component $\mathbf{h}\cdot\mathbf{nn}$ contributes so that we replace $\mathbf{h}$ by this component. Noting that the trace of a product of two operators is independent on their order we immediately get Equation 34 with

$$I_0 P = \mathrm{Tr}'\,(g\dagger\mathbf{h}\cdot\mathbf{n} + g\mathbf{h}\dagger\cdot\mathbf{n})/(2s_i + 1) \qquad 38.$$

It should be noted that the result Equation 34, or Theorem 2, depends on the time reversal argument which allows us to drop the $\mathbf{h}\times\mathbf{h}\dagger$ term in Equation 37. It follows that Theorem two may hold even when the process is not an elastic scattering provided the polarizer and analyzer represent inverse reactions. We cannot use Theorem two when the polarizer is an inelastic scattering from a nucleus, however, since the inverse reaction would require starting with an excited nucleus.

Triple scattering,—(75, 82, 83). Further information about the matrix M may be obtained from triple scattering experiments. Such experiments are designed to determine how the second scatterer changes the direction or magnitude or both of the polarization of the proton; thus, the first scatterer serves simply as a polarizer and the final scatterer as an analyzer.

If we apply Equation 31 to the second scattering to relate the polarization of the outgoing nucleon beam $\langle\boldsymbol{\sigma}\rangle_f$ to that of the incoming nucleon beam $\langle\boldsymbol{\sigma}\rangle_i$ the most general form of the relation will be

[6] We shall not prove here the remainder of Theorem 3, which is a simple extension of the theorem of Yang and others on the angular distribution for the unpolarized cases (81).

$$I_2\langle\vec{\sigma}\rangle_f = I_{02}\{[P_2 + D\langle\vec{\sigma}\rangle_i\cdot\mathbf{n}_2]\mathbf{n}_2 + [A\langle\vec{\sigma}\rangle_i\cdot\mathbf{k}_2 + R\langle\vec{\sigma}\rangle_i\cdot(\mathbf{n}_2\times\mathbf{k}_2)]\mathbf{s}_2 + [A'\langle\vec{\sigma}\rangle_i\cdot\mathbf{k}_2 + R'\langle\vec{\sigma}\rangle_i\cdot(\mathbf{n}_2\times\mathbf{k}_2)]\mathbf{k}_2'\} \quad 39.$$

where the coefficients P_2, D, etc., are arbitrary functions of the scattering angle and the energy. The P_2 term comes from letting S^ν on the right hand side of Equation 31 equal the unit matrix; if the initial beam is unpolarized, P_2 is the only term and $I_2 = I_{02}$, thus the equation reduces to Equation 11b. The other terms correspond to S^ν equalling one of the components of the incident spin operator. The proof that Equation 39 is the most general form is based on the requirement that the right-hand side transform like an axial vector, which rules out terms like $\langle\vec{\sigma}\rangle\cdot\mathbf{s}_2\mathbf{n}_2$, etc. The triple-scattering experiments thus involve five new parameters D, A, R, A', and R'. From time-reversal arguments it may be shown that only four of these are independent: for an infinitely heavy target they are related by $R' = -A$, while for the case of two particles of equal mass (non-relativistically)

$$(A + R')/(A' - R) = \tan(\theta/2) \quad 40.$$

Since polarization along the direction of motion cannot be detected by a single scattering, the third scattering is chosen to detect either $\langle\vec{\sigma}\rangle_f\cdot\mathbf{n}_2$ or $\langle\vec{\sigma}\rangle_f\cdot\mathbf{s}_2$. The corresponding asymmetries in triple scattering are designated e_{3n} and e_{3s}, defined as $[I_3(+) - I_3(-)]/[I_3(+) + I_3(-)]$, where I_3 $(\pm)$ corresponds to $\cos\phi_3 = \pm 1$ ($\cos\phi_3 = \mathbf{n}_2\cdot\mathbf{n}_3$ for e_{3n}; $\cos\phi_3 = \mathbf{s}_2\cdot\mathbf{n}_3$ for e_{3s}). If the beam incident on the second scatterer follows a single scattering the initial polarization is along $\mathbf{n}_1$, and thus perpendicular to $\mathbf{k}_2(=\mathbf{k}_1')$. In this case I_2 is given by Equation 35. From Equation 39 we now see that only D and R can be determined from a simple succession of three scatterings:

(1) D experiment. If the third scattering is selected to measure e_{3n} we have from Equations 3, 35 and 39

$$e_{3n} = P_3\langle\vec{\sigma}\rangle_f\cdot\mathbf{n}_2 = P_3(P_2 + DP_1\cos\phi_2)/(1 + P_1P_2\cos\phi_2) \quad 41.$$

To measure D we choose $\cos\phi_2 = 1$ (or -1), which means that all three scattering planes are parallel (Fig. 2). In this case if the initial beam is completely polarized, that is $P_1 = 1$,

$$\langle\vec{\sigma}\rangle_f = \mathbf{n}_2(P_2 + D)/(1 + P_2)$$

The parameter D is seen to give the extent to which the second scattering depolarizes the beam since a value of D equal to unity is required for there to be no depolarization. Actually the direction of polarization may even be reversed, since it is possible for D to be less than $(-P_2)$. It is easily shown that D is limited in general by

$$-1 + 2|P_2| \leqq D \leqq 1$$

For the case of a spinless target D equals unity, as may be seen by setting θ_u equal to zero in Equation 8.

(2) R experiment. If the third scattering is selected to measure e_{3s} we have

$$e_{3s} = P_3\langle\overline{\sigma}\rangle_f \cdot \mathbf{s}_2 = P_3RP_1 \sin\phi_2/(1 + P_1P_2\cos\phi_2) \qquad 42.$$

To measure R we choose $\sin\phi_2 = 1$, which means that the successive scattering planes are at right angles to each other (Fig. 2), in particular, the second scattering must be up (or down) with respect to the first scattering plane. It is easily shown that R is limited by $|R| \leqq (1-P_2^2)^{1/2}$.

It is possible, in addition, to determine A if a magnetic field directed perpendicular to $\mathbf{n}_1$ and $\mathbf{k}_1'$ is placed between the first and second scatterers. The effect of the magnetic field H is to bend the particle (upward or downward with respect to the plane of the first scattering) through an angle δ and simultaneously to turn the spin through an angle Δ, which is larger than δ because of the anomalous magnetic moment of the nucleon. Thus the spin is turned with respect to the direction of motion by an angle (75, 84):

$$\begin{aligned}\Delta - \delta &= (\mu - z)(eH/mc)t \\ &= (\mu - 1)E\delta \text{ for protons} \qquad 43.\end{aligned}$$

where mc^2E is the total proton energy (including rest energy). In particular, it would be possible to turn the spin so that it pointed along the direction of motion before the second scattering (Fig. 2). Similarly the parameter R' may be determined by placing a magnetic field between the second and third scatterer. To determine A' two magnetic deflections would be necessary. Thus successions of three scatterings, some with magnetic deflections interposed, can determine completely all the parameters of Equation 39; nothing more can be learned from an additional number of scatterings.

Correlation experiments.—To gain further information about the matrix M we must consider cases of Equation 31 in which either $\langle\overline{S^\nu}\rangle_i$ or $\langle\overline{S^\mu}\rangle_f$ involves the target spin as well as the incident-particle spin. This means either that the target is polarized or that one measures the polarization of the recoiling target most likely in correlation with the polarization of the outgoing nucleon. Some examples are the possible correlation experiments for the case of nucleon-nucleon scattering with an unpolarized target and with no magnetic fields (83, 85). For the particle scattered at angle θ one can measure the component $\langle\overline{\sigma_t}\rangle$ along the directions $\mathbf{n}$ or $\mathbf{s}$ perpendicular to $\mathbf{k}'$. The recoiling target (indicated by subscript t) has a direction $\mathbf{k}_t'$ and one can measure its polarization $\langle\overline{\sigma}\rangle$ along the directions $\mathbf{n}$ or $\mathbf{s}_t$ $(=\mathbf{n}\times\mathbf{k}_t')$. For nucleon-nucleon scattering we have, using the non-relativistic kinematics, $\mathbf{k}' = \mathbf{s}_t = \mathbf{P}$ and $\mathbf{k}_t' = -\mathbf{s} = -\mathbf{K}$. In the polarization-correlation experiment we are interested in the dyadic $\langle\overline{\sigma\sigma_t}\rangle_f$ of which we can measure the $\mathbf{n}\,\mathbf{n}$, $\mathbf{KP}$, $\mathbf{Kn}$, and $\mathbf{nP}$ components. Considering only these four components and restricting ourselves to components of the incident spin $\langle\overline{\sigma}\rangle_i$ perpendicular to the incident direction the most general expression is

$$I_2\langle\overline{\sigma\sigma_t}\rangle_f = I_{02}(C_{nn} + C_{nn}{}^p\langle\overline{\mathfrak{G}}\rangle_i\cdot\mathbf{n}_2)\mathbf{n}_2\mathbf{n}_2 + I_{02}(C_{KP} + C_{KP}{}^p\langle\overline{\mathfrak{G}}\rangle_i\cdot\mathbf{n}_2)\mathbf{K}_2\mathbf{P}_2 + I_{02}\{C_{Kn}{}^p\langle\overline{\mathfrak{G}}\rangle_i\cdot(\mathbf{n}_2\times\mathbf{k}_2)\}\mathbf{K}_2\mathbf{n}_2 + I_{02}\{C_{nP}{}^p\langle\overline{\mathfrak{G}}\rangle_i\cdot(\mathbf{n}_2\times\mathbf{k}_2)\}\mathbf{n}_2\mathbf{P}_2$$

where the coefficients C_{nn}, etc. are arbitrary functions of θ and energy. Terms like $\langle\overline{\mathfrak{G}}\rangle_i\cdot\mathbf{n}_2\mathbf{K}_2\mathbf{n}_2$ are ruled out since $\langle\mathfrak{G}\mathfrak{G}_2\rangle$ is even under inversion of the coordinates. Thus there are two distinct experiments of this type starting with an unpolarized beam and four more using a polarized beam.

Relativistic effects.—The present treatment in terms of Pauli spinors has been nonrelativistic. The polarization of a free Dirac particle, however, may be defined by means of the Pauli spinor in the rest frame of the particle (4). A relativistic version of the formalism, developed by Stapp (85), then reduces to equations very similar to those given in this paper. Stapp shows that the nonrelativistic formalism gives correct equations if the targets may be considered as infinitely heavy or if all the scatterings are in one plane. Relativistic corrections (besides the usual relativistic kinematics) are needed, however, in equations for R, A, C_{KP}, etc., for nucleon-nucleon scattering.

5. Nucleon-Nucleon Scattering

It is well known that the nucleon-nucleon interaction has a fairly complicated spin dependence, indicated in the neutron-proton system at low energies by the difference between the singlet and triplet ground states and by the quadrupole moment of the deuteron. It is hard to determine the nature of the spin-dependent interaction effective in scattering simply from a measurement of the differential cross section of an unpolarized beam since one then averages over all initial and final spin states. The various polarization experiments described in Section 4 have proved very useful in obtaining more information.

p-p Scattering formalism.—For proton-proton scattering the amplitude M is a 4×4 matrix in the composite spin space. Invariance arguments (77, 78) show that only five of the sixteen matrix elements are independent and that the most general form of the matrix may be written

$$M = BS + C(\mathfrak{G}+\mathfrak{G}_t)\cdot\mathbf{n} + \tfrac{1}{2}G(\mathfrak{G}\cdot\mathbf{K}\mathfrak{G}_t\cdot\mathbf{K} + \mathfrak{G}\cdot\mathbf{P}\mathfrak{G}_t\cdot\mathbf{P})T + \tfrac{1}{2}H(\mathfrak{G}\cdot\mathbf{K}\mathfrak{G}_t\cdot\mathbf{K} - \mathfrak{G}\cdot\mathbf{P}\mathfrak{G}_t\cdot\mathbf{P}) + N\mathfrak{G}\cdot\mathbf{n}\mathfrak{G}_t\cdot\mathbf{n}T \qquad 44.$$

where S and T are projection operators which restrict the term in question to singlet and triplet states, respectively. The five amplitudes B, C, G, H, and N depend only on the scattering angle θ for any one energy. As a consequence of the identity of the protons, one may show that B, H, and C are even functions of $\cos\theta$ whereas G and N are odd functions. At each scattering angle $\theta\leqq90°$ we might hope to determine experimentally nine real quantities, the magnitudes of the five amplitudes and their relative phases. Nine experimental results at any one angle will give us nine quadratic equations

in these nine unknowns. The resulting ambiguity may thus still be very large; on the other hand, it may turn out that a smaller number of experiments already puts strong limitations on these amplitudes. The following are some of the possible experiments listed roughly in order of increasing difficulty: (*a.*) Unpolarized cross section $I_0(\theta)$; (*b.*) Polarization $P(\theta)$; (*c.*) Triple scattering in one plane ($\theta \leqq 90°$): $D(\theta)$; (*d.*) Triple scattering: successive planes at right angles ($\theta \leqq 90°$): R(θ); (*e.*) Triple scattering with magnetic field before second scattering ($\theta \leqq 90°$): $A(\theta)$; (*f.*, *g.*) Correlation experiments with unpolarized beam: C_{nn}, C_{KP}; (*h.* to *j.*) D, R, and A for $\theta \geqq 90°$; (*k.*)(*l.*) Triple scattering with magnetic field after second scattering ($\theta \leqq 90°$ and $\theta \geqq 90°$): $R'(\theta)$; (*m.* to *p.*) Correlation experiments with polarized beam. Since for a proton emerging in a direction (θ, ϕ) there is a recoil proton in a direction $(\pi-\theta, \pi+\phi)$, the differential cross-section $I(\theta, \phi)$ must equal $I(\pi-\theta, \pi+\phi)$. It follows that $I_0(\theta) = I_0(\pi-\theta)$ and that $P(\theta) = -P(\pi-\theta)$. The minus sign arises from the fact that the contribution of the polarization to the cross section is proportional to $P(\theta) \cos \phi$ (Equation 35). In the case of a triple-scattering experiment, however, the results for $\theta < 90°$ and $\theta > 90°$ are independent since the two emerging protons need not have the same polarization. If the final state were a pure triplet (or a pure singlet) state there would be no difference between these two polarizations, and the triple scattering experiments would be related by $D(\theta) = D(\pi-\theta)$, $R(\theta) = R'(\pi-\theta)$, and $A(\theta) = -A'(\pi-\theta)$. The failure of these equalities is a measure of singlet-triplet interference. (Expressions for most of these experimental quantities in terms of the amplitudes may be found in refs. 80, 83, and 85.)

Because of the short-range character of the nuclear forces, it should be possible to describe the scattering in terms of a relatively small number of phase shifts. It is therefore unnecessary to determine nine experimental quantities at one angle since the experiments at different angles are not really independent. (Expressions for the amplitudes and experimental quantities in terms of phase shifts are given in Refs. 75, 85 to 87.)

p-p Polarization experiments.—The scattering of polarized protons from hydrogen to determine $P(\theta)$ has been performed at a number of energies between 140 Mev and 430 Mev (20 to 24, 88 to 90); the results are summarized in Figure 7. The polarization is particularly interesting when viewed in conjunction with the differential cross section which is approximately constant at 3.7 millibarns/steradian independent of angle and energy in this region. Thus at 140 Mev the isotropic cross section could have been explained by singlet S-wave scattering alone, but the sizeable polarization observed requires that at least about half of the scattering comes from triplet states. Furthermore, even at 140 Mev the polarized cross section I_0P does not vary as $\sin \theta \cos \theta$ so that from Equation 36 the triplet scattering is not simply due to p-waves but requires some contribution from f-waves. It is possible to fit all the results for $P(\theta)$ using the first two nonvanishing terms of Equation 36

$$I_0P = (a_1 \cos \theta + a_2 \cos^3 \theta) \sin \theta \qquad 45.$$

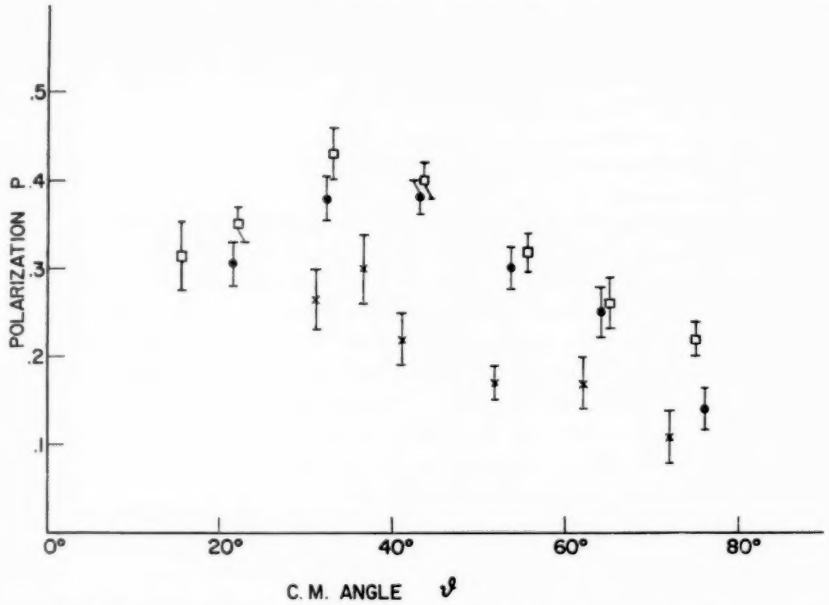

FIG. 7. Polarization P vs. center of mass angle θ for p-p scattering. Crosses—140 Mev (24, 88). Black circles—310 Mev (90). Squares—415 Mev (23). (In using the data of Refs. 23 and 24, the beam polarization has been taken as .53 and .64, respectively, rather than the values given in these references. These new values have been communicated by the authors.)

with the coefficients in mb/ster. given by[7] (88, 89, 23)

140 Mev $a_1 = 1.11 \pm .12$	$a_3 = 1.28 \pm .20$
310 Mev $a_1 = 2.0 \pm .3$	$a_3 = 1.6 \pm .4$
415 Mev $a_1 = 1.6 \pm .3$	$a_3 = 3.0 \pm .5$

This large polarization is the first evidence for the noncentral character of the proton-proton triplet state interaction. The flatness of the differential cross section can also be explained in part by the noncentral triplet interaction; thus, for example, effective spin-orbit coupling in ${}^3P$ scattering leads to $\sin {}^2\theta$ terms in the cross section in addition to the usual $\cos {}^2\theta$ terms.

For a triple-scattering experiment it is necessary that the scattered proton have an energy well above 100 Mev in order that its polarization can be easily analyzed (cf. Fig. 6). Consequently until now such experiments have

[7] The value at 140 Mev is obtained (88) from all the data up to March, 1956, only some of which are shown in Figure 7. At the higher energies there is some indication of an a_5 term. Thus while the data can be fit satisfactorily without such a term, a somewhat better fit might be one in which a_3 is smaller and a_5 is almost as big as a_3. Errors quoted are estimated from the statistics of the experiment and do not include errors due to the possibility of an a_5 term.

not been performed below 300 Mev. The most complete information on proton-proton scattering comes from the Berkeley group which has measured D, R and A at 310 Mev at angles between 20° and 80° in the center-of-mass system (90, 91, 92). Measurements of D have also been made at 420 Mev. (93). Further information (94, 95) on the proton-proton system at high energies comes from measurements (96, 97) of the asymmetry of the mesons produced in the reaction $p+p \rightarrow \pi^+ + d$ using a polarized beam.

A variety of attempts have been made to analyze the results of proton-proton scattering experiments. The most ambitious procedure is to try to determine the scattering from a static potential model. It has been shown that qualitative agreement with polarization can be obtained using either a tensor force or a direct spin-orbit coupling for the noncentral interaction (98), but no potential has been found that gives a quantitative fit to the data. This last fact would appear to have little significance, since the ratio of the number of potentials for which calculations have been made to all conceivable potentials is undoubtedly of measure zero.

A much less ambitious but still quite difficult program is to try to determine the phase shifts from the data (85, 88, 92, 99 to 104). At an energy around 100 Mev one might hope that only four phase shifts need be considered, the singlet S phase shift δ_0 and the three p-wave phase shifts δ_1^0, δ_1^1, δ_1^2, where the superscript indicates the total angular momentum J. Four pieces of data that could be used to obtain these are the two coefficients in the nuclear differential cross section (which is of the form $a+b\cos^2\theta$), the polarization (that is, a_1 in Equation 36) and the nuclear-Coulomb interference in the cross section. Unfortunately the lowest energy for which polarization measurements have been made is 140 Mev, at which energy there is definite evidence for f-waves. Making as good as fit as possible ignoring the f-waves, Taylor (88) finds at 140 Mev: $\delta_0 = 22°$, $\delta_1^0 = -40°$, $\delta_1^1 = -10°$, $\delta_1^2 = 7°$. Feshbach & Lomon (103) find very similar results from a somewhat different approach except they take δ_1^1 equal to 0°. These results can be changed drastically if d- and f-waves are included (102). At 310 Mev a phase-shift analysis has been made including ^1S, ^3P, ^1D, and ^3F waves; this makes eight phase-shifts plus a mixing parameter representing the coupling between the $^3P_2$ and $^3F_2$ states. The large amount of data available has made it feasible to try to determine all of these nine parameters; four distinct sets of phase shifts have been found which give good fits to the data (92). All these sets turn out to have small values for the F phase shifts, indicating that it may well be proper to ignore higher partial waves at this energy. One result of interest has been the negative value of the singlet S phase-shift in qualitative agreement with the repulsive core models of nuclear potentials.

Equation 45 should not apply at small angles where Coulomb interference effects may be observed. These effects have been calculated by Garren (105), who points out that additional information for the determination of phase-shifts could be obtained by accurate small-angle polarization measurements. It is of interest to note that the magnetic moment interaction of the proton plays an important role in this effect.

n-p Polarization experiments.—For the case of neutron-proton scattering the most general form of $M(\theta, \phi)$ is given by Equation 44 supplemented by a term with a spin dependence $(\boldsymbol{\sigma} - \boldsymbol{\sigma}_t) \cdot \mathbf{n}$, which clearly cannot occur for identical particles. We adopt the convention that $\boldsymbol{\sigma}_t$ is the proton spin and $\boldsymbol{\sigma}$ is the neutron spin. The number of distinct polarization experiments is larger for *n-p* scattering than for *p-p* because we must distinguish the cases in which the neutron is polarized from those in which the proton is polarized. Thus there are two distinct double-scattering experiments: (*a*) scattering of a polarized neutron beam from hydrogen, and (*b*) scattering of a polarized proton beam from target neutrons. The cross section for the first of these could be given by Equation 32 while the second would require replacing $\boldsymbol{\sigma}$ in Equation 32 by $\boldsymbol{\sigma}_t$. It is immediately evident that these two can be different only if M is not symmetric in $\boldsymbol{\sigma}$ and $\boldsymbol{\sigma}_t$; thus the difference is entirely due to the $(\boldsymbol{\sigma} - \boldsymbol{\sigma}_t) \cdot \mathbf{n}$ term in M. According to the hypothesis of charge symmetry, the scattering amplitude M must not change when proton and neutron are interchanged and thus this term must equal zero and the two double-scattering experiments become identical. Both of these experiments have been performed at about the same energy: (*a*) the Carnegie Tech group (106) has measured the left-right asymmetry of recoil protons using a 16 per cent-polarized beam of neutrons with an effective energy of 350 Mev; (*b*) the Berkeley group (90, 107) has scattered their 310-Mev polarized proton beam from deuterium and measured the left-right asymmetry of the recoil neutrons (or of the scattered protons counted in coincidence with the recoil neutrons). The similarity of the results of these two experiments may be considered as evidence for charge symmetry.

If we further assume charge independence we may write the matrix M for *n-p* scattering

$$M_{np} = \tfrac{1}{2}(M_0 + M_1)$$

where M_T is the scattering for the two-nucleon system with total isotopic spin T and each M_T can be expanded separately in the form of Equation 44. For *p-p* scattering we have simply $M_{pp} = M_1$. If we look at any *n-p* scattering result, in particular, $(I_0P)_{np}$, we can consider it as the sum of three terms: (*a*) terms due to $T=1$ states alone, (*b*) terms due to $T=0$ states alone, and (*c*) interference terms between $T=1$ and $T=0$ states. The first of these may be determined directly from *p-p* scattering. The last two terms are distinguishable by the fact that the second involves interference between states of the same parity (e.g., ^3S and ^3D), whereas the third involves interference between states of opposite parity (e.g., ^3S and ^3P). As a consequence the second term contributes terms to I_0P which are odd with respect to 90° whereas the third contributes even terms. We are thus able to isolate the value of I_0P for pure $T=0$ scattering (108):

$$(I_0P)_{00}(\theta) = 2(I_0P)_{np}(\theta) - 2(I_0P)_{np}(\pi - \theta) - (I_0P)_{pp}(\theta) \qquad 46.$$

The results for *n-p* scattering at 310 Mev have been fitted (107) to Equation 36:

$$(I_0P)_{np} = \sin\theta\{ - .34 + .45\cos\theta + 1.30\cos^2\theta + 2.93\cos^3\theta\}\text{mb/ster.}$$

where the coefficients have large unstated errors. Combining this result with Equation 46 and Equation 45 at 310 Mev we obtain

$$(I_0P)_{00} = 10 \sin\theta \cos^3\theta \text{ mb/ster} \qquad 47.$$

While this result is subject to sizeable errors, the following conclusions seem justified: (*a*) there is definite evidence for an appreciable non-central interaction effective in $T=0$ states at 300 Mev; one might expect that this is related to the tensor force which shows itself in $T=0$ states at low energies; (*b*) there is a considerable contribution to *n-p* scattering at 310 Mev from *D* states or higher; and if we ignore higher states, the ${}^3D_3$ state must be quite important. Note that the result of Equation 47 cannot be obtained from ${}^3D-{}^3S$ interference nor from ${}^3D_1-{}^3D_2$ interference.

Very interesting preliminary results on *n-p* polarization have been obtained at 100 Mev (88, 109). These also have been fitted to Equation 36:

$$(I_0P)_{np} = \sin\theta\,(1.1 \pm 0.2 + [3.5 \pm .7]\cos\theta) \text{ mb/ster.} \qquad 48.$$

If we assume $(I_0P)_{pp}$ at 100 Mev is less than at 140 Mev it contributes little to Equation 46 so that we are satisfied with making a reasonable guess as to its value. We then obtain at 100 Mev

$$(I_0P)_{00} = 13 \sin\theta \cos\theta \text{ mb/ster.}$$

This result is most likely too high and is certainly very rough but it indicates important non-central interaction effects in the $T=0$ scattering at 100 Mev. It seems most reasonable to ascribe this primarily to ${}^3S-{}^3D$ interference (103, 104).

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EXCITATION OF NUCLEI BY CHARGED PARTICLES[1]

By N. P. Heydenburg and G. M. Temmer

Department of Terrestrial Magnetism, Carnegie Institute of Washington, Washington, D.C.

I. INTRODUCTION

In this review we shall be concerned with a discussion of the excitation of atomic nuclei to discrete (bound) low-lying[2] energy levels by the Coulomb field of moving charged particles. The occurrence of this type of process was claimed as early as thirty-five years ago (1) and was discussed by Rutherford *et al.* (2) and Landau (3) around 1930. Although some indications were reported before the war for appreciable inelastic scattering at energies where compound-nucleus formation would be very unlikely (4, 5), it was not until 1953 that incontrovertible evidence for the excitation of nuclei by the purely electromagnetic interaction between passing charged particles and the nuclear protons was reported (6, 7).

A clear-cut distinction can and should be made between electron and heavy-particle excitation. The former seems to have been successful to date only with rather high-energy electrons (of the order of 100 Mev or more) in a limited number of the lighter nuclei; heavy-particle excitation, usually referred to as Coulomb excitation or electric excitation (mainly protons, deuterons, and alpha particles) on the other hand, has been successfully applied over most of the periodic table ($_9F^{19}$ to $_{94}Pu^{239}$) and has revealed a considerable number of new properties of low-lying nuclear excited states, especially as to their systematic behavior upon passing from one nuclear species to the next. The relative simplicity of these experiments both in their execution and in their interpretation, has given us, in the method of Coloumb excitation, one of the most powerful tools for the study of many properties of stable nuclei.

Since a very simple relationship, based only on detailed balancing arguments, exists between upward and downward electromagnetic transitions which connect two discrete nuclear states (absorption and emission), the method of Coulomb excitation supplements and augments the scope of the more conventional methods used in determining the properties of electromagnetic transitions in nuclei. For an excellent review of the latter subject we refer the reader to an earlier volume in this series (8), where other references may be found.

There are essentially two methods for the detection of this inelastic scattering process: (*a*) detection of the inelastic group or groups of particles, (*b*) detection of the radiation emitted from the excited nuclear state or

[1] The survey of the literature pertaining to this review was concluded in March, 1956.

[2] By "low-lying" we shall generally mean levels having of the order of, or less than 1 Mev excitation energy.

states resulting from the inelastic encounter. The latter method can be further subdivided into two categories: (i) detection of gamma radiation emitted in the deexcitation, and (ii) detection of the internal conversion electrons emitted from various atomic shells in the transition. All of these approaches have already been used successfully, and will be discussed in some detail below.

After a brief discussion of electron excitation, we shall describe the present status of Coulomb excitation experiments, and some of the implications of the results obtained from these experiments.

II. EXCITATION OF NUCLEAR BOUND STATES BY ELECTRONS

As implied in the introduction we shall not include in this review the electro-disintegration of nuclei, i.e., the excitation of nuclei by electrons which leads to the subsequent emission of one or more nucleons. That subject is closely related to the field of photonuclear reactions and is more properly discussed in that connection.

Since the interaction between electrons and nuclei is almost entirely of the electromagnetic type,[3] and since the theory of this interaction is comparatively well understood, electrons constitute a powerful probe of the nuclear interior.[4]

A. Low-Energy Electrons

Very little definite evidence exists on the direct excitation of nuclear energy levels by low-energy electrons, i.e., electrons having energies of the order of the level energies. Most of this work was performed by the group at the Notre Dame electrostatic electron generator at energies below 2.5 Mev (14). The technique invariably used in these experiments was to turn the beam off after bombardment, and to observe the residual activity of a conveniently long-lived isomeric state, resulting from cascade transitions from one or more (unobserved) higher-lying states which are excited in the primary event.[5] The uncertainty in the interpretation of the results stems from the fact that the cross section for the excitation of these higher states by photons is about two orders of magnitude larger than that by electrons, and the latter inevitably produce bremsstrahlung in the target. The observation of prompt gamma radiation excited by low-energy photon or electron bombardment has apparently not been attempted. A number of theoretical papers dealing with the interpretation of such experiments can be found in the literature (15, 16, 17).

[3] We can neglect the very small nonelectromagnetic electron-nucleon interaction in these considerations.

[4] μ-mesons, both bound and free, having vanishingly small nuclear interaction, also constitute a useful electromagnetic probe; for a discussion of both theory and experiment bearing on this aspect of μ-mesons, see (9–13).

[5] The direct excitation of the isomeric transition is always negligible because of its high multipolarity and long lifetime.

High-Energy Electrons

Much has been learned concerning radial nuclear charge density distributions [e.g., (13)] from the comprehensive elastic scattering experiments of the Stanford linear accelerator group using high-energy electrons in the 100 to 500 Mev region. In a few of the lighter nuclei these workers have been able to observe inelastic electron groups as well, and to determine their angular distributions (18 to 21). Schiff (22) has pointed out that such experiments can reveal the radial dependence of the nuclear transition charge density. The most extensively studied nucleus so far is C^{12}, where three (known) excited states at about 4.4, 7.7, and 9.6 Mev have been observed through their inelastic groups, in addition to the elastic scattering peak. This is illustrated in Figure 1 for 80-degree scattering and 187 Mev (19). The cross section for

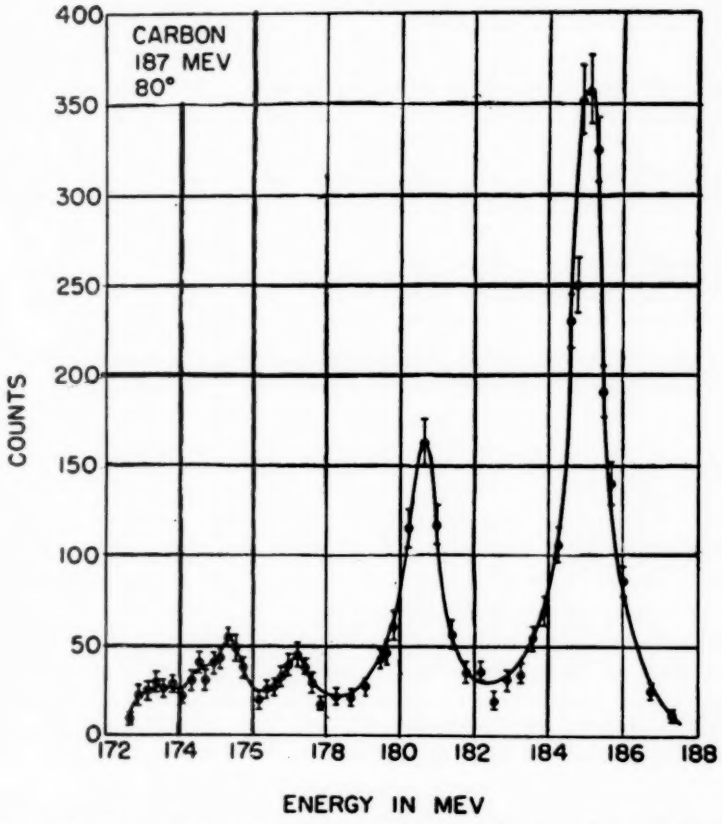

Fig. 1. Elastic and inelastic scattering of 187-Mev electrons from carbon at 80° to the beam. Largest peak to the right at 185 Mev corresponds to elastic scattering; next largest peak at 181 Mev represents inelastic scattering to the 4.43-Mev 2^+ state in C^{12}; small peaks at 177 and 175 Mev correspond to inelastic scattering to the 7.6 Mev and 9.5 Mev states in C^{12}, respectively. [Taken from Fregeau & Hofstadter (19).]

the excitation of the strongest inelastic group (the one leading to the first-excited state of 2^+ character at 4.43 Mev) at 90 degrees to the incident beam remains almost constant from 80 Mev to 187 Mev at a value of about 10^{-31} cm.^{2}/steradian while the elastic peak decreases by a factor of about 400 between these electron energies, becoming equal to the inelastic differential cross section at 187 Mev. An explanation of this striking behavior has been attempted (22a). It thus becomes practically impossible, even with the rather high available resolution ($\sim$0.5 per cent) to detect inelastic peaks at electron energies much below about 80 Mev. This is mainly because of the appreciable bremsstrahlung "tail" always present on the low energy side of the elastic electron peak.

The second excited state of C^{12} at 7.65 Mev is believed to have character 0^+ (like the ground state), and hence the transition induced by the electrons must be of the electric monopole or $E0$ type. Because of the transverse nature of the electromagnetic field, no one-photon process carrying off zero angular momentum is allowed, and the transition can only occur by a "longitudinal photon" interaction taking place within the confines of the charge and current distribution, i.e., inside the nucleus. Electrons (and μ-mesons) are thus privileged in being able to electrically excite such monopole transitions. Their special role in shedding light on certain features of nuclear structure has recently been discussed (23, 24). Other levels in the lighter nuclei excited by high-energy electrons have also been reported (18, 20, 21). Attempts to detect the characteristic deexcitation gamma radiation under high-energy electron bombardment have thus far not been successful (25).

III. EXCITATION OF NUCLEAR BOUND STATES BY HEAVY CHARGED PARTICLES (COULOMB EXCITATION; ELECTRIC EXCITATION)

The development of the energy sensitive sodium iodide scintillation counter, more than any other single fact, is undoubtedly responsible for the discovery of Coulomb excitation three years ago. There are, in principle, no new facts concerning electromagnetic transitions which are revealed exclusively in Coulomb excitation; the reasons for the superiority of the method in most instances are mainly to be found in its simplicity, its ability to measure lifetimes the better the shorter they are, and the possibility of reaching excited states which, because of accidents of nature, are either inaccessible or have transitions which are unobservably weak following radioactive decay from neighboring isobars. A further advantage is the uniformity with which the properties of each nuclear species can be determined irrespective of the extraneous factors which inevitably accompany investigations using the conventional methods of beta- and gamma-ray spectroscopy. Of course one is limited in that only stable or near stable nuclear species lend themselves as targets for these experiments, and that one of the states involved in the primary transitions observed must, of necessity, be the ground state.

The conventional Van de Graaff generator has so far turned out to be the most satisfactory source of particles for observing Coulomb excitation.

In fact, because of unfavorable background conditions, the effect has only been observed in a few isolated instances under cyclotron bombardment, and then only by the observation of delayed isomeric gamma radiation (5, 26) (in a way analogous to that described in Section II A above), or by bombardment with triply-charged N^{14} ions of 26 Mev at Oak Ridge (27), and of 15.6 Mev at Leningrad (27a). By far the most extensive work has been reported with Van de Graaff protons and alpha particles; deuterons have also been used on occasion when detecting conversion electrons (28, 29).

Whereas electrons interact with nuclei only via the electromagnetic field, heavy charged particles exert both electric and nuclear forces on target nuclei; in order to benefit from the simplicity of the former, and to avoid confusion inherent in our lack of understanding of the latter, one takes advantage of the short range nature of the nuclear interaction and tries to keep the energy of the bombarding particle below the Coulomb barrier. This will minimize penetration and consequent formation of a compound nucleus by the capture of the incident particle. This of course is not to imply that Coulomb excitation is unimportant for energies near the top or even above the barrier; in fact, since two barrier penetrations are involved, of which one takes place at reduced energy (outgoing particle), the probability of direct Coulomb excitation may outweigh the compound-nuclear inelastic scattering leading to the same excited state until the bombarding energy reaches the top of the barrier. There is, of course, always an effective maximum angle of deflection which decreases with increasing bombarding energy and within which Coulomb excitation always predominates (36). However, in the interest of simpler interpretation of the results, and to avoid experimental difficulties, it is best to limit the bombarding energy to values well below the Coulomb barrier.

A. Coulomb Excitation Theory

Total cross section.—The theory of this process was considerably ahead of conclusive experiments for many years (3, 30 to 36); because of the technical limitations on measuring gamma-ray transitions efficiently and accurately, most of the early theoretical work was directed toward the prediction of indirect effects susceptible of possible measurement, such as deuteron break-up by the electric field of a nucleus, inelastic deuteron scattering, and the like (30, 31, 33). The first detailed, semi-classical calculation of the Coulomb inelastic cross section was due to Ter-Martirosyan (36). He assumed classical, hyperbolic trajectories for the bombarding particles, i.e., he assumed that

$$\eta \equiv \frac{Z_1 Z_2 e^2}{\hbar v} \gg 1 \qquad 1.$$

where Z_1e and Z_2e are the charges of projectile and target nucleus, respectively,[6] and v is their relative velocity; the nucleus, however, was viewed as

[6] Throughout this article, subscript 1 on any symbol shall refer to the projectile, and subscript 2 to the target nucleus.

a quantum-mechanical system capable of undergoing discrete transitions. Condition 1 merely insures the régime of geometrical optics, i.e., that the wavelength be short compared to the size of the "object," which in this case is of the order of the distance of closest approach. Ter-Martirosyan calculated the electric dipole ($E1$) and electric quadrupole ($E2$) excitation cross sections, specifically for the case of negligibly small energy of excitation ($\Delta E \rightarrow 0$). Although the $E1$ cross section had previously been expressed in closed form (30, 31), the $E2$ cross section which, as we shall see, turns out experimentally to be most important, is not amenable to such a treatment. Alder & Winther (37) numerically evaluated the semi-classical cross section associated with the $E2$ component in the electric field of the incident particle. A number of investigators soon began to tackle the question of the validity of the semiclassical approximation for various experimental situations; a numerical, quantum-mechanical calculation for a specific situation and no energy loss (38, 39) revealed the essential adequacy of the semi-classical result for that limiting case; a more general treatment, but still for $\Delta E = 0$, showed the relation between the quantum-mechanical and semi-classical results as a function of the parameter η (40). A number of numerical comparisons have also been made between the semi-classical and quantum-mechanical approach for finite energy loss (41 to 44). Because of the long range character of the Coulomb force, an enormous number of partial waves contribute in an essential way to the process [up to 250 in some cases, see reference (47)]; hence, complete quantum-mechanical calculations for finite energy loss ΔE have only recently been carried out with the aid of electronic computers (45 to 49). They show that as far as the total inelastic Coulomb cross section is concerned, a slightly modified form of the simple semi-classical expression (37, 50), stemming from a WKB approximation calculation (51), adequately describes the process to a high degree of accuracy (better than one per cent) over most of the range of excitation and bombarding energies usually encountered. The correct $E2$ cross section is given by (45)

$$\sigma(E2) = \frac{m_1^2 v_f^2}{Z_2^2 e^2 \hbar^2} B(E2) f_{E2}(\eta_i, \xi), \qquad 2.$$

where m_1 is the reduced mass of the projectile, v_f is the relative velocity after collision, $Z_2 e$ is the charge of the target nucleus, $B(E2)$ is the so-called reduced (upward) transition probability (52), and the function $f_{E2}(\eta_i, \xi)$ is dimensionless and has been numerically evaluated (47, 49). It depends on two dimensionless parameters: η_i, which is the Sommerfeld number defined in equation 1, for the initial relative velocity v_i, and $\xi = \eta_f - \eta_i$, where η_f contains the final relative velocity v_f. In the limit of vanishing energy transfer ΔE the collision parameter ξ approaches the value

$$\xi(\Delta E \rightarrow 0) \simeq \frac{Z_1 Z_2 e^2}{\hbar v} \cdot \frac{\Delta E}{2E}. \qquad 3.$$

This quantity is easily seen to be the ratio of the time of passage of the projectile to the nuclear period associated with the transition ΔE. For small values of ξ ($\xi \ll 1$) the function f_{E2} is approximately independent of ξ and of order unity, whereas for values ξ of 0.5 or greater, the function f_{E2} decreases exponentially with increasing ξ, falling off by about an order of magnitude for every $\Delta\xi$ of about 0.5 (see Table I). It is useful to keep in mind the combina-

TABLE I

THE CLASSICAL TOTAL CROSS SECTION FUNCTION $f_{E2}(\infty, \xi)$ FOR $E2$ EXCITATION VS. ξ*

ξ	$f_{E2}(\infty, \xi).10^{+P}$	P
0.0	0.8954	0
0.1	0.8638	0
0.2	0.7289	0
0.3	0.5608	0
0.4	0.4046	0
0.5	0.2781	0
0.6	0.1844	0
0.7	0.1189	0
0.8	0.7511	1
0.9	0.4663	1
1.0	0.2855	1
1.2	0.1035	1
1.4	0.3628	2
1.6	0.1238	2
1.8	0.4143	3
2.0	0.1363	3

* From Alder & Winther (45).

tion of physical quantities occurring in equation 3, as well as the rough trends mentioned above, for the purpose of estimating magnitudes of the Coulomb excitation cross section.

In the classical limit, i.e., when $\eta_i \to \infty$ the function $f_{E2}(\infty, \xi)$ becomes the classical function $g_2(\xi)$ (37) obtained by integration along the hyperbolic orbit of the projectile, except for a constant factor, i.e., we have $f_{E2}(\infty, \xi) = (2\pi^2/25)g_2(\xi)$. We tabulate the classical limit of f_{E2} in Table I. The situation for finite values of η_i is illustrated in Figure 2 in terms of the ratio to the limiting classical function of Table I; more complete tabulations of the function $f_{E2}(\eta_i, \xi)$ can be found in the literature (47, 49).

We should like to emphasize here that all these considerations are completely independent of any assumed nuclear model, and rest entirely on the well established properties of the radiation field and the Coulomb interaction. It is precisely this property of the Coulomb excitation process which

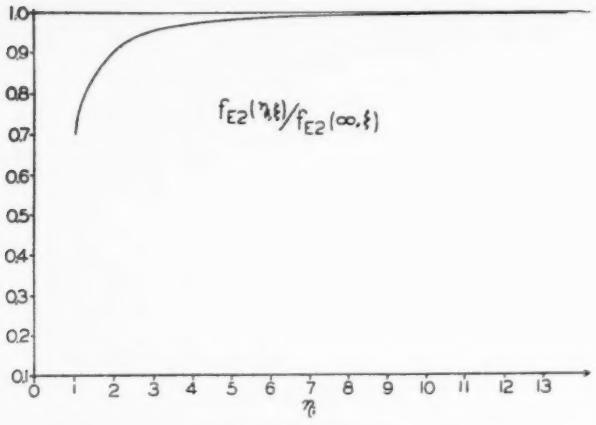

FIG. 2. Ratio of quantum-mechanical Coulomb excitation function $f_{E2}(\eta_i, \xi)$ to its semi-classical limit $f_{E2}(\infty, \xi)$ (see Table I) as a function of the Sommerfeld parameter $\eta_i = Z_1 Z_2 e^2/\hbar v_i$. [Taken from Alder & Winther (45).]

accounts for its great usefulness in the determination of the intrinsic nuclear quantity B, the reduced transition probability (see below under *Excitation probability*). Various nuclear models will make definite predictions as to the magnitude of the quantity B, as we shall see later, and comparison with experiment can be conveniently made at this level. The same quantity B can also be obtained from lifetime measurements (8, 52) and resonance fluorescence experiments (53, 54), except that one must remember the statistical spin factor which relates the reduced transition probabilities between states with spins I_i and I_f as follows:

$$B(i \to f) = B(f \to i)\frac{2I_f + 1}{2I_i + 1}. \qquad 4.$$

Some theoretical attention has been paid to the calculation of the higher electric multipole components as well as the magnetic multipoles, but no clear-cut experimental evidence for these much less probable components has as yet been found. For the sake of completeness, we list the general expressions for the total Coulomb excitation cross sections for both electric and magnetic multipoles (50):

$$\textit{Electric } 2^\lambda\textit{-pole: } \sigma(E\lambda) = a^2\left(\frac{Z_1 e^2}{\hbar v_i}\right)^2 \frac{B(E\lambda)}{e^2 a^{2\lambda}} \left(\frac{v_f}{v_i}\right)^{2\lambda-2} f_{E\lambda}(\eta_i, \xi) \qquad 5.$$

$$\textit{Magnetic } 2^\lambda\textit{-pole: } \sigma(M\lambda) = a^2\left(\frac{Z_1 e^2}{\hbar c}\right)^2 \frac{B(M\lambda)}{e^2 a^{2\lambda}} \left(\frac{v_f}{v_i}\right)^{2\lambda-1} f_{M\lambda}(\eta_i, \xi) \qquad 6.$$

where

$$a = \frac{Z_1 Z_2 e^2}{m_1 v_i^2}$$

is one-half the distance of closest approach, and all other quantities have been defined previously. Aside from the intrinsic quantities B, the most important factors in these expressions are the functions f which vary strongly with the parameter ξ. Exact, quantum-mechanical calculations for f are available only for $E1$ (30, 31, 47, 55) and $E2$ (47, 49) Coulomb excitation at present. However, semi-classical approximations to these functions have been computed and are numerically tabulated for $E1$, $E2$, $E3$, $E4$, $M1$, and $M2$ excitation (50) in the limit of large η; these tabulations should be adequate for most actual conditions likely to be encountered. A forthcoming review article (56) will include formulae for these more unusual cross sections. The formalism for the quantum-mechanical computation of higher multipole moments is available (45, 48).

We should like to emphasize that the exact quantum-mechanical calculations mentioned earlier were performed under non-relativistic conditions, as well as under the assumption that all phenomena connected with the possible barrier penetration of the projectile can be neglected. They further neglect effects of retardation (long wave length approximation) and the lack of coincidence between the center of mass and the center of the multipole expansion; no satisfactory treatment of the latter problem exists at present (48, 55). These are not too serious restrictions on the general applicability of the results, but some of them will need to be revaluated, for instance for the interpretation of experiments on light nuclei or with heavy bombarding particles, or those revealing some resonant yield along with Coulomb excitation (57 to 60).

Some theoretical work has recently appeared concerning magnetic dipole ($M1$) and mixed ($M1$–$E2$) Coulomb excitation (56, 61). An inherent difference exists between protons and alpha particles as far as $M1$ is concerned because the proton possesses a magnetic moment. As far as the total cross section is concerned, no interference occurs between $M1$ and $E2$ excitation, and the two contributions merely add; however, the $M1$ component is usually negligible owing to the relatively low velocity of the projectiles (for experimental evidence, see below), and we need only take the $E2$ cross section into account, even though an $M1$–$E2$ mixture may be allowed *a priori*. Of course, once a state is excited, it will decay via its appropriate $M1$–$E2$ mixture, the $M1$ component being usually predominant. We shall elaborate on these matters in the section on *Multipolarity* below.

Angular distribution of inelastic particles.—Extremely good energy resolution is required to resolve the inelastic groups resulting from Coulomb excitation, from the elastic group due to Rutherford scattering, (separated by the order of 100 kev) there being of the order of one inelastic event in several thousand elastic scatterings, for representative values of ξ and η (see above), and at favorable backward angles. Some promising preliminary results have been obtained with a high-resolution magnetic spectrograph and 7 Mev protons incident on several rare earth nuclei (56, 62) and on gold (56, 63). Further work is needed to decide whether these conditions insure the exclu-

sive régime of Coulomb excitation. Semiclassical calculations to determine the angular distribution of Coulomb inelastic groups are available in graphical form (50). In contrast to the case of electron excitation, there seems to be little of intrinsic nuclear interest to be learned from such angular distributions which cannot be obtained from either total cross section measurements or angular distribution measurements of gamma radiation following the excitation.

Angular distribution of gamma radiation following Coulomb excitation.— This problem bears a close formal analogy to the problem of directional correlation of cascade radiation [see e.g. (64)] or the angular distribution of radiation from oriented nuclei [see e.g. (65)]; in all these cases the first event (i.e., beam direction in Coulomb excitation, emission of the first quantum, spatial alignment of a nucleus) serves to establish an axis of quantization with respect to which the succeeding radiation is emitted with a characteristic angular distribution. Each one of these particular cases, however, requires some special modification, the one for Coulomb excitation being the most elaborate. If we write the directional correlation of two successive gamma quanta emitted from the nuclear level sequence $I_0 \rightarrow I_1 \rightarrow I_2$ as

$$W(\theta) = \sum_n A_n P_n (\cos \theta) \qquad 7.$$

we can write the related expression for the angular distribution of gamma rays following Coulomb excitation as

$$W(\theta) = \sum_n a_n(\eta_i, \xi) A_n P_n(\cos \theta). \qquad 8.$$

The summation extends only over even Legendre polynomials, the highest term being given by the smallest numerical value among $2L_1$, $2L_2$, and $2I_1$, where L_1 and L_2 are the multipolarities of the two transitions. The coefficients A_n have been put in a convenient form (66) involving products of two factors, each of which depends only on a single step of the transition. Extensive tabulations of these factors are available (64, 66, 67), covering all values of the parameters likely ever to be encountered in practice (67). The particle parameters $a_n(\eta_i, \xi)$ which are independent of intrinsic nuclear properties, must be obtained from Coulomb excitation theory. The spin sequence as written above applies to Coulomb excitation if I_0 refers to the ground state spin of the target nucleus, I_1 to the excited state spin, and I_2 to the final state spin, which is very often the ground state spin again. For the case of $E2$ excitation (most important case) we merely consider the first transition $I_0 \rightarrow I_1$ to take place by pure $E2$ radiation, the second step $I_1 \rightarrow I_2$ occurs by a mixture of $M1$ and $E2$ radiation in general, unless $|I_1 - I_2| = 2$, in which case it must also be of pure $E2$ character. For the general $E2$ excitation case expression 8 will have the form

$$W(\theta) = 1 + a_2(\eta_i, \xi) A_2 P_2(\cos \theta) + a_4(\eta_i, \xi) A_4 P_4(\cos \theta). \qquad 9.$$

The first calculation of a_2 and a_4 was again made on a semi-classical basis (37, 50) and unfortunately contained an error in sign which bedeviled the

field for some time. This error, independently committed in the work of Biedenharn & Class (40), has now been cleared up (45 to 49, 68 to 71a). Exact quantum-mechanical calculations are now available for the coefficient a_2 encountered in $E1$ excitation (47, 55), as well as for the coefficients a_2 and a_4 occurring in $E2$ excitation (47, 49). No other angular distribution coefficients have been calculated to date. Unlike the situation in the total excitation cross section discussed above, considerable dependence on η of the coefficients a_2 and a_4 exists in the quantum-mechanical formulation, and the semi-classical value is a poor approximation. For example, at $\xi=0.5$, the coefficient a_2 for $\eta_i=2$ is about 30 per cent larger than the classical value ($\eta_i=\infty$), while the coefficient a_4 for $\eta_i=2$ is about four times larger than its semi-classical limit. It is worth noting that a_2 can take on values greater than unity; hence observed anisotropies[7] in Coulomb excitation can exceed the corresponding γ-γ correlation values (66). a_4 is mostly negative and of the order 0.2. The anisotropies always increase with increasing ξ (e.g. decreasing projectile energy).

As in ordinary γ-γ correlation extranuclear effects may attenuate the angular distribution of gamma rays if the intermediate state lifetime exceeds values of the order of $\sim 10^{-9}$ second [see e.g. (64)] (depending to a considerable degree on the type and strength of the interaction), and have been found in F^{19} (72); in fact, by applying an external magnetic field a determination of the nuclear g-factor of the 197-kev excited state was made (73, 74). A similar attempt is under way for one of the even-even rare earth nuclei (75).

Polarization of Coulomb-excited gamma rays.—In analogy to the case of the linear polarization-direction correlation in a γ-γ cascade, we expect linear polarization in Coulomb-excited gamma radiation. The theoretically predicted strength and angular distribution of polarization can be obtained by carrying over the formalism for direction polarization correlation (64, 66), to the Coulomb excitation case, following the prescription outlined above for the case of the angular distribution. This will again involve the coefficients a_2 and a_4 (56). Some very recent experiments (75a) have determined the polarization in a number of cases and have shown how such measurements can be used to remove the inherent ambiguity which often exists in the determination of the mixture ratio δ from the angular distribution of gamma radiation (see equation 15 below; also Section IIIB).

Coulomb-excited gamma rays should constitute a useful source of polarized gamma radiation not requiring coincidence detection.

Particle-gamma ray directional correlation.—The possibility exists of detecting the inelastic particle in coincidence with the gamma radiation from the excited state, thus relaxing somewhat the requirement for high resolution of the inelastic group. With resolving times of $\sim 10^{-8}$ second, such an experiment becomes marginal for a favorable situation.

[7] We define the anisotropy as $\epsilon=[W(O)/W(\pi/2)]-1$.

Some calculations have been performed which predict certain small deviations (of the order of a few per cent) from the *a priori* expected particle-gamma-ray direction correlation (76), the effect being proportional to $Z_1^{1/2}m_1^{1/2}$ (for fixed distance of closest approach a); heavy particle bombardment should therefore be preferable. This effect is caused by the finite amplitude for the Coulomb excitation process and the consequent possibility of redistributing the population among the magnetic substates of the excited state during the time of passage of the projectile. The effect allows one, in principle, to determine the static electric quadrupole moment Q of an excited state. It is furthermore sensitive to the sign of Q, and hence represents one of the few methods for the determination of the sign of the quadrupole moment (11, 12), even for nuclei whose spin is zero in the ground state.

For a very detailed and complete exposition of Coulomb excitation theory, we refer to the review article of Alder *et al.* (56).

B. General Features of Coulomb Excitation Experiments

Bohr & Mottelson pointed out even before the discovery of Coulomb excitation the great usefulness of this process in detecting those features in nuclei which involve cooperative, many-particle motion (35); the fact that cross sections for excitation turn out generally not to be of the single particle size but up to 100 times larger, explains both the recent experimental success and early theoretical pessimism concerning Coulomb excitation. Before turning to the interpretation of some of these experiments, we shall discuss a number of the more immediate and practical considerations which are needed for their evaluation.

Detection.—We have already mentioned the general methods of detecting Coulomb excitation in the introduction. The detection of inelastic (heavy) particle groups, which has so far been of limited success, was briefly mentioned above. By far the most universally used method has been the detection of gamma radiation, mostly by means of scintillation counters (6, 7, 57 to 60, 72, 77 to 105), and proportional counter (106). The detection of internal conversion electrons, carried out exclusively by means of a type of beta-ray spectrometer with shaped magnetic field invented by Kofoed-Hansen (107), has had important successes mainly in the rare earth and heavy-element region where the conversion coefficients for low-lying levels are large (29, 108 to 111). A spectrometer such as used by Huus and collaborators (29, 108), is shown in Fig. 3a, while a typical gamma-ray detection setup is shown in Fig. 3b (57). A conversion electron spectrum for Ta^{181} (137-kev transition) as obtained with 3.45 Mev alpha particles is shown in Fig. 4a (109); a spectrum of gamma-rays Coulomb-excited by 6-Mev alpha particles in four even isotopes of palladium is illustrated in Fig. 4b (92).

It is interesting to reflect that Coulomb excitation could have been discovered twenty years earlier using electron detection, since no recent advances in technique, such as occurred in gamma-ray spectroscopy, were required. The energy resolution achieved in conversion spectrometry is gen-

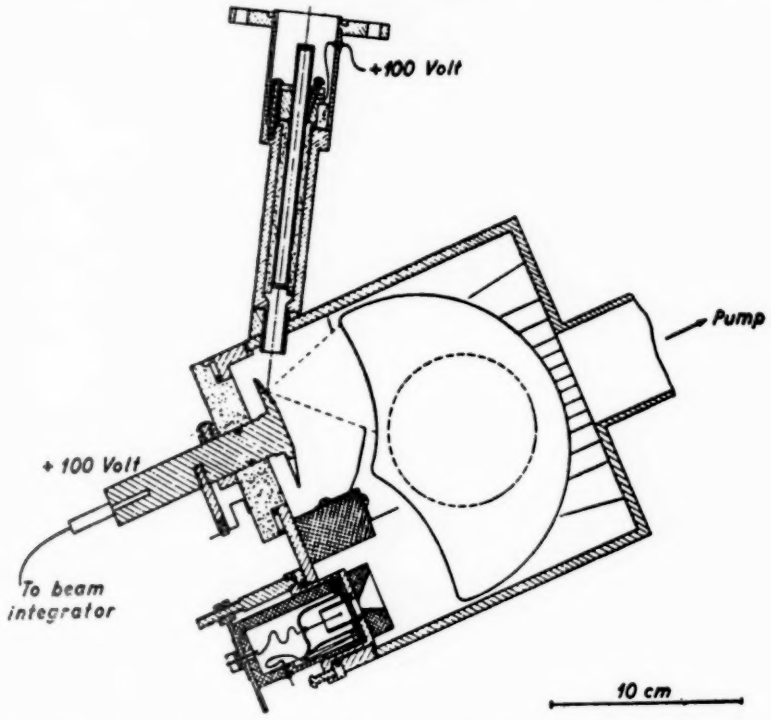

FIG. 3(a). Typical equipment for the detection of Coulomb excitation. (*a*) Internal conversion spectrometer as used by Huus & Zupančič (108); (*b*) scintillation crystal spectrometer as used by Heydenburg & Temmer (57).

erally superior to the scintillation method. There is however an important difference in efficiency between the gamma-ray and conversion-electron methods: for transitions of the order of 100 kev to 200 kev, the efficiency (including solid angle) for the former might approach 50 per cent, whereas the latter, depending to some extent on the desired energy resolution, is a few per cent at best. In addition, gamma-ray work is usually carried out with (infinitely) thick targets, while electron targets must of necessity be limited to the order of 100 micrograms per cm.^{2}; this accounts for an additional, considerable loss in intensity. For transition energies much in excess of 200 kev the smallness of the conversion coefficients makes gamma detection the uncontested means for observing Coulomb excitation.

Targets.—A few words are in order concerning the choice of target material and thickness. Pure metallic elements are, of course, most desirable for any incident particle; however, oxides and other compounds must sometimes be used of necessity, especially with isotopically enriched targets and rare earth oxides. The presence of oxygen leads to certain undesirable effects with

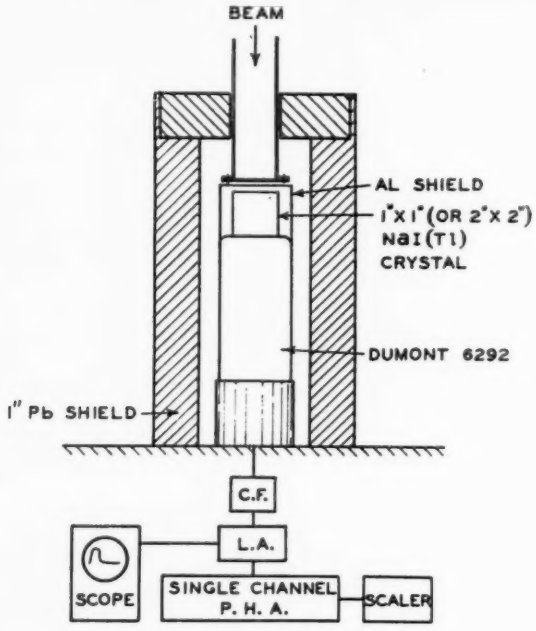

FIG. 3(b). See legend on 3(a).

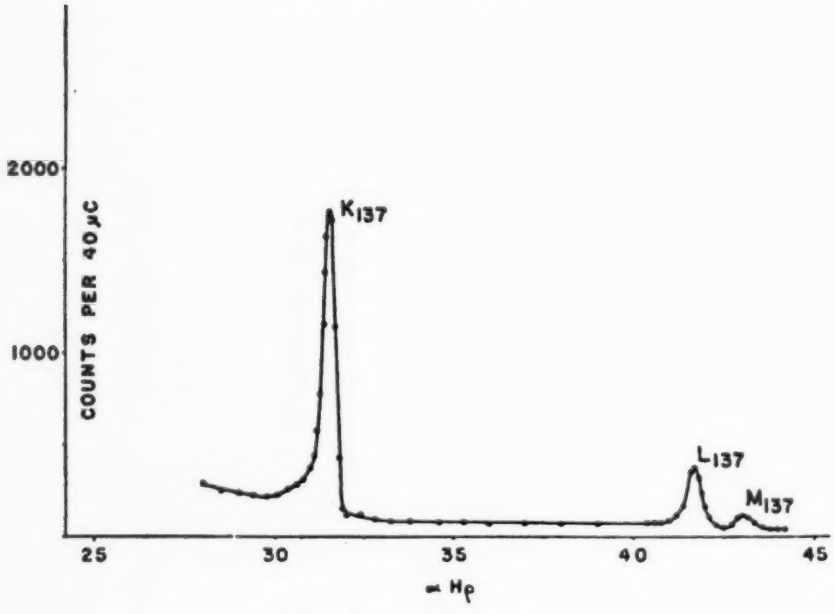

FIG. 4(a). See legend on 4(b).

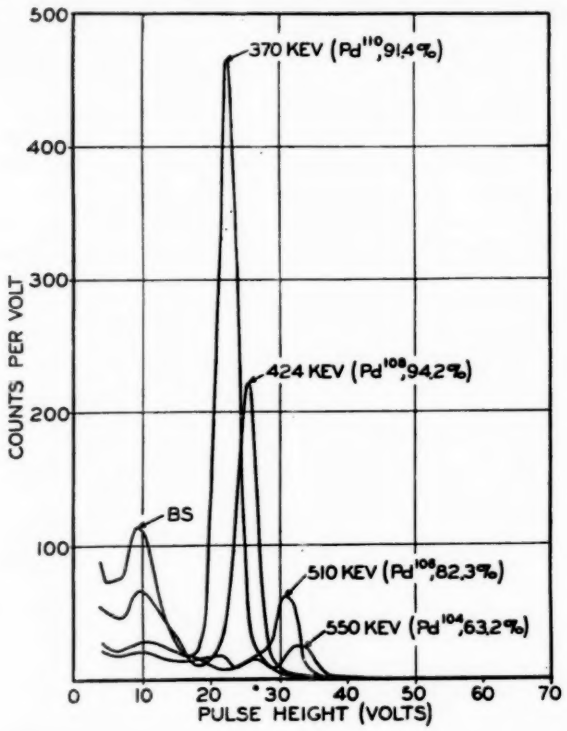

FIG. 4(b). Typical spectra observed in Coulomb excitation. (*a*) Internal conversion lines from *K*, *L*, and *M* shells due to 137-kev transition in Ta^{181}, excited by 3.45-Mev alpha particles; observed by Bernstein & Lewis (109); (b) scintillation spectra of four even-even nuclei of Pd (isotopically enriched targets as indicated) observed by Temmer & Heydenburg (92). Peak marked BS is due to backscattered radiation.

protons (60, 91) and to a lesser extent, with alpha particles.[8] In addition, one must of course caution against radiation from impurities, the most important being fluorine and sodium.[9]

When one wishes to determine the absolute radiation yield in order to determine the total (thin target) Coulomb excitation cross section, one faces different problems with thin and thick targets. With a thin target, the exact

[8] The only major effect ascribable to the presence of oxygen, aside from an intensity loss for all radiations because of target dilution, is the appearance of a 342-kev gamma ray from the $O^{18}(\alpha, n\gamma)Ne^{21}$ reaction (58, 92, 98).

[9] Gamma rays at 109 and 197 kev, and at 446 kev, are often indicative of the presence of traces of F^{19} or Na^{23}, respectively. Since each of these elements simultaneously also emits higher energy gamma radiation [following either (p, α) or (α, p) reactions], we have found that a ratio comparison will usually resolve the dilemma of assignment of gamma rays at these energies.

thickness will have to be determined, either by the well-known resonance shift method (112) plus a knowledge of the stopping power, or more directly by the elastic (Rutherford) scattering of the bombarding particles (29). Corrections are then still to be applied to equation 2 for the finite target thickness. With an infinitely thick target, we need to integrate the expression for the Coulomb excitation cross section over the track length of the slowing down incident particle. Because of the rapid variation of the cross section (equation 2) with ξ (equation 5), and hence with bombarding energy, the main contribution to the yield will come from the beginning of the track, so that a certain insensitivity to details of the stopping power variation with energy results. We have generally favored thick targets for absolute yield determinations, even in the few cases where we obtained sufficient yield for thin-target experiments. The advantage of the thick-target method for the comparison of several isotopes of one element, or even of elements not greatly differing in atomic number, should be readily apparent. While it is desirable to integrate the cross section over stopping power exactly, and there is a deplorable lack of universal applicability about these integrations, a much more rapid and, we believe, quite adequate method, suggested by Huus & Mottelson (113), will enable one to make rapid comparisons of different conditions for thick-target situations. We believe it worth while to outline this approach [see also references (29) and (56)]. We assume that $\Delta E \ll E$, i.e., that equation 3 holds, and hence that no distinction has to be made between v_i and v_f. These conditions are adequately fulfilled for bombarding energies greater than $\sim$4 Mev and excitation energies below about 300 kev. We now make the following assumptions concerning the dependence of the stopping power on particle energy and the atomic number Z_2 of the target: (*a*) the stopping power, dE/dx, varies as $E^{-1/2}$; (*b*) the stopping power per atom varies as $Z^{1/2}$. These assumptions are motivated by the experimental determinations on stopping powers at low energies (114 to 118). Because of the above mentioned steepness of the function $f_{E2}(\xi)$ the thick-target yield is quite insensitive to assumption (*a*), as we shall see. Assumption (*b*) is not essential, and empirical values can be used for any given element. If we denote by a subscript 0 those quantities referring to the incident particle energy, and if we express all variables in equation 2 in terms of ξ, we obtain the following expression for the $E2$ thick-target yield:

$$Y = N\sigma(E_0)\left[\frac{E_0 y(\xi_0)}{(dE/dx)_0}\right], \qquad 10.$$

where $y(\xi_0)$ stands for an integral given by

$$y(\xi_0) = 2/3\xi_0^{5/3}\frac{1}{f_{E2}(\xi_0)}\int_{\xi_0}^{\infty}\frac{f_{E2}(\xi)}{\xi^{8/3}}d\xi, \qquad 11.$$

and N is the number of target atoms per cc. The factor in brackets occurring in equation 10 clearly represents the equivalent target thickness. $\sigma(E_0)$ is given by equation 2. In Table II we list some representative numerical values of the function y (ξ_0) for a series of values of ξ_0.[10]

[10] We are indebted to T. Huus for furnishing these values.

TABLE II

THE THICK-TARGET FUNCTION $y(\xi_0)$ *versus* THE PARAMETER ξ_0. $y(\xi_0)$ IS DEFINED IN EQUATION 11 (113).

ξ_0	$y(\xi_0)$	ξ_0	$y(\xi_0)$
0.0	0.400	1.0	0.0903
0.1	0.332	1.2	0.0768
0.2	0.260	1.4	0.0674
0.3	0.213	1.6	0.0602
0.4	0.178	1.8	0.0579
0.6	0.135	2.0	0.0540
0.8	0.108		

It turns out that if we had used an energy-independent stopping power instead of assumption (*a*), the value of $y(0)$, where the largest target penetration (and hence the maximum discrepancy) is to be expected, would have been 0.500 instead of 0.400 (see Table II). Hence small deviations from the $1/v_1$ law should be of little consequence. We have compared the exact integration with the simplified approach given above in a few cases, and found agreement within a few per cent. Furthermore, excellent agreement between experimental thick-target excitation curves and the prescription given above has been found over considerable ranges in ξ (see Fig. 8 on p. 98).

Multipolarity of Coulomb excitation.—With the sole exception of the 109-kev first-excited state of F^{19}, which has been shown to be reached by a pure electric-dipole transition ($\frac{1}{2}^+ \rightarrow \frac{1}{2}^-$) (72, 119), all of the nearly 200 transitions induced by Coulomb excitation to date are believed to be of the electric quadrupole type, that is to say, they connect the ground state with excited states of the same parity, and spins I^* obeying the selection rule $\rho \leqq |I_0 - I^*| \leqq 2$, where $\rho = 2$, 1, or 0, according to whether $I_0 = 0$, $\frac{1}{2}$, or $> \frac{1}{2}$. Let us limit our considerations for the moment to a comparison of the $E1$ and $E2$ modes of excitation. In view of what is generally known about nuclei (and about Coulomb excitation) it is not too surprising to find this overwhelming preponderance of $E2$ transitions. Now the *a priori* $E1$ excitation probability is, for a typical case, some 300 times larger than $E2$ excitation (similar factors apply for successively higher multipolarities). However, there are two main reasons against the observation of these transitions: (*a*) the scarcity of states of opposite parity within a few hundred kilovolts of the ground state, and (*b*) the extremely small dipole transition matrix elements connecting these rare states [$B(E1)$ about $10^{-3} - 10^{-7}$ of the single-proton estimate; see Fig. 3 of reference (8)]. Recently a number of low-lying states of character 1^- has been found in some heavy, even-even nuclei (120), decaying by pure $E1$ radiation to the 0^+ ground states; some evidence of low-energy $E1$ transitions to the ground states of odd-A transuranic nuclei (121) and rare earth nuclei (122) has also been reported. No evidence of Coulomb excitation of any of these states has been found as yet (98), which is in keeping with con-

sideration (*b*) above: the smallness of $B(E1)$ more than overcomes the favorable intrinsic $E1$ Coulomb excitability.[11]

The situation for nuclei with an even number of protons and an even number of neutrons is quite clear. Since all of them have character 0^+ in their ground states (no known exception) and character 2^+ in their first excited states (123, 124),[12] these states can always be reached by $E2$ Coulomb excitation, and essentially all of them have been so excited.

Most of the nuclei with odd mass number have low-lying excited states which can be reached by magnetic dipole transitions ($\Delta I = 0$ or 1, no change in parity). In fact, practically all such transitions which are Coulomb-excited (via $E2$) return to the ground state with a characteristic radiation mixture containing a preponderant constituent of $M1$ radiation, as is to be expected from single-particle considerations. Significant deviations from this behavior will be discussed later on. Unfortunately there exists no case of a pure $M1$ transition which does not admit of $E2$ excitation also.[13] However, in two cases of very small $E2$ admixture (La^{139} and Pr^{141}) no evidence of any Coulomb excitation was found in spite of conditions comparing favorably as to excitation energy with neighboring nuclei (98). In addition, several cases with measurable $E2$ components but predominant $M1$ matrix elements show that $M1$ Coulomb excitation is unimportant (58). This merely bears out what is expected theoretically in view of the depressing influence of the retardation factor v/c for (relatively) slow particles and magnetic transitions on the one hand, and what is known empirically for the size of the quantity B ($M1$) on the other hand (8).

Finally, there is some evidence as to the (expectedly) negligible contribution of $E3$ Coulomb excitation even for a favorable case. There exists a well-known $E3$ isomeric transition of 17.5 second half life and 160 kev energy in Se^{77}. This isomer was observed in Coulomb excitation (100), but it was found that the excitation took place by cascade from a 244-kev level, the latter being reached by $E2$ transition. Although only about two per cent of the ground-state return from the 244-kev level went via the isomer, we find that this indirect mode of excitation (by $E1$) is still about 100 times more prolific than any direct excitation calculated from the value of $B(E3)$ known from the lifetime. Similar conclusions can be reached from some work on silver (85).

Bjerregaard & Huus (28) have described an ingenious means (suggested by Bohr & Mottelson) for determining the multipolarity of a given transition, making use only of very general properties of the Coulomb excitation theory. They verified the $E2$ nature of the known $E2$ transition in even-even

[11] This is especially true relative to the enhanced $E2$ transitions which occur in the regions where these $E1$ transitions are reported (see Strongly deformed nuclei below).

[12] The only known exceptions are: O^{16}, Ca^{40}, Ge^{72}, Zr^{90} (all 0^+), and $Pb^{208}(3^-)$.

[13] The only exceptions are the pure $M1$ transitions in the odd-odd nuclei N^{14} (2.31 Mev) and RaE (Bi^{210}, 46 kev) connecting states of equal parity and spins 0 and 1, respectively.

tungsten nuclei by bombardment with protons, deuterons, and alpha particles. The method merely requires a measurement of the ratio of the relative thin-target yields for a given transition, with two projectiles having different values of Z_1/A_1, at bombarding energies chosen so as to insure equal values of the parameter ξ. Making use of the cross section as given by equation 5, the function $f_{E\lambda}$ drops out and we compute the yield ratio, e.g., for alpha particles and protons at equal ξ (taking the classical limit and neglecting reduced mass effects for simplicity) to be

$$\frac{\sigma_\alpha(\xi)}{\sigma p(\xi)} \cong 4 \times 2^{2\lambda/3} = \begin{cases} 6.35 \text{ for } \lambda = 1 \ (E1) \\ 10.1 \ \text{ for } \lambda = 2 \ (E2) \\ 16.0 \ \text{ for } \lambda = 3 \ (E3) \end{cases} \qquad 12.$$

where $E_\alpha(\xi) \cong 2.52 E_p(\xi)$. These ratios can be computed more accurately, but will show the general behavior given above, so that a relatively crude experiment ($\sim$20 per cent) will suffice to decide between the different "quantized" possibilities. This method has been successfully applied to the following odd-A nuclei and transitions: Mn^{55}, 128-kev state (58), F^{19}, 197-kev state, and Na^{23}, 446-kev state (59). They all yield ratios near 10 ($E2$ exciation), as expected. It is interesting to discuss the cases of F^{19} and Na^{23}, where, at the proton energies appropriate for this experiment, strong resonances exist. It was possible to show in both cases that the residual, inelastic excitation yield between resonances is just the amount expected from $E2$ Coulomb excitation. This situation is illustrated in Figure 5 which shows the thin-

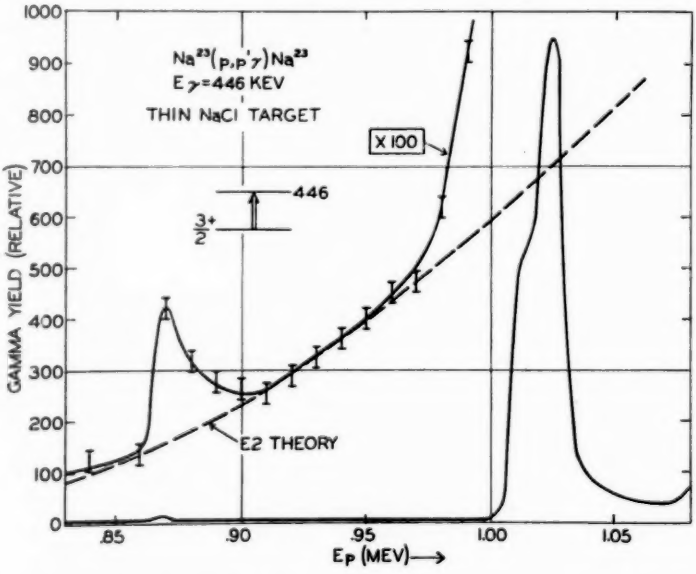

FIG. 5. Inelastic excitation of 446-kev state in Na^{23} by protons. Thin NaCl target. Residual yield between compound-nuclear resonances accounted for by $E2$ Coulomb excitation. [Taken from Temmer & Heydenburg (59).]

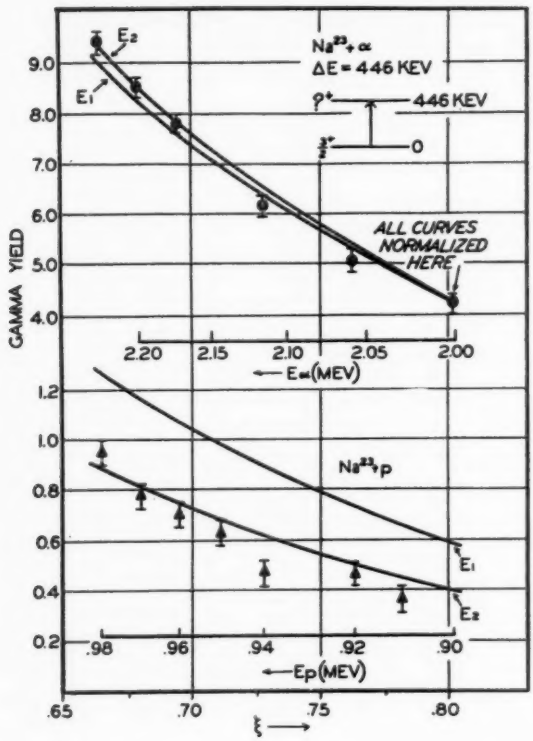

FIG. 6. Proof of $E2$ nature of the excitation of 446-kev level in Na^{23}. Comparison of thin-target yields due to alpha particles and protons. Note common scale of ξ and different energy ranges for alphas and protons (increasing to the left). Only the difference between predicted $E1$ and $E2$ curves in the lower half of the figure is significant. All four curves normalized as indicated. [Taken from Temmer & Heydenburg (59).]

target yield of 446-kev radiation versus proton energy. The corresponding energy range for alpha particles shows smooth behavior (see Fig. 7 below). Figure 6 shows a plot of relative yields *vs.* a common scale of ξ. All curves are normalized at one point, and represent a number of independent determinations of the yield ratio. Only the spread in the lower part of the figure between $E1$ and $E2$ curves is significant.

Excitation functions.—The first experimental checks of semi-classical Coulomb excitation theory, bearing on the correct ξ-dependence of the function $f_{E2}(\xi)$ revealed satisfactory agreement for thick targets (6, 77, 86, 87); for a number of the stronger transitions in lighter nuclei it was possible to obtain excitation curves with thin targets (58). One of these curves for Na^{23} is illustrated in Figure 7. Insufficiently thin targets are revealed by their

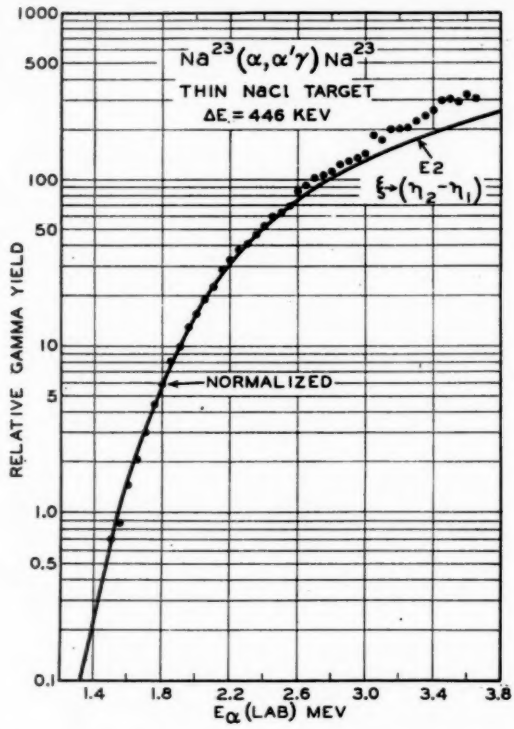

FIG. 7. Thin-target Coulomb excitation curve of 446-kev level for alpha particles on Na^{23}. Solid curve is theoretical semi-classical *E*2 excitation function. Note smooth variation in the alpha-energy range used for the comparison in Figure 6. [Taken from Temmer & Heydenburg (58).]

steeper excitation functions; the ultimate test of a thin target is to observe no further change in shape of the excitation curve upon using a thinner target, i.e., one yielding less radiation. The modifications introduced into the simple, semi-classical expression by the WKB approximation (51) were definitely needed to improve the agreement with the observed shapes of the excitation functions (58). However, no evidence for the type of behavior illustrated in Figure 2 for low values of η_i was found within the accuracy of these experiments. The departures above the theoretically expected *E*2 behavior shown in Figure 7 at the higher energies are to be ascribed to contributions from nuclear inelastic scattering of alpha particles in which a compound state is formed (i.e., penetration effects which are not considered in the theory; see above). A thick-target curve (see Targets above) covering a more than 20,000-fold yield increase and showing excellent agreement with *E*2 theory is shown in Figure 8 for Ru^{104} which has the lowest excited state and largest transition probability of any even-even nucleus below A = 145.

This sort of agreement can usually be taken as confirmation of the $E2$ character of the excitation process, although sometimes the distinction between the shapes of $E1$ and $E2$ excitation is small over limited ranges of bombarding energy (58, 90). To sum up, then, the experimentally observed excitation curves confirm the theoretically predicted dependence upon ξ to within the experimental error (about 3 to 5 per cent) over a considerable range of ΔE (50 kev to 850 kev), Z_2 (9 to 94), and E (1 Mev to 7 Mev).

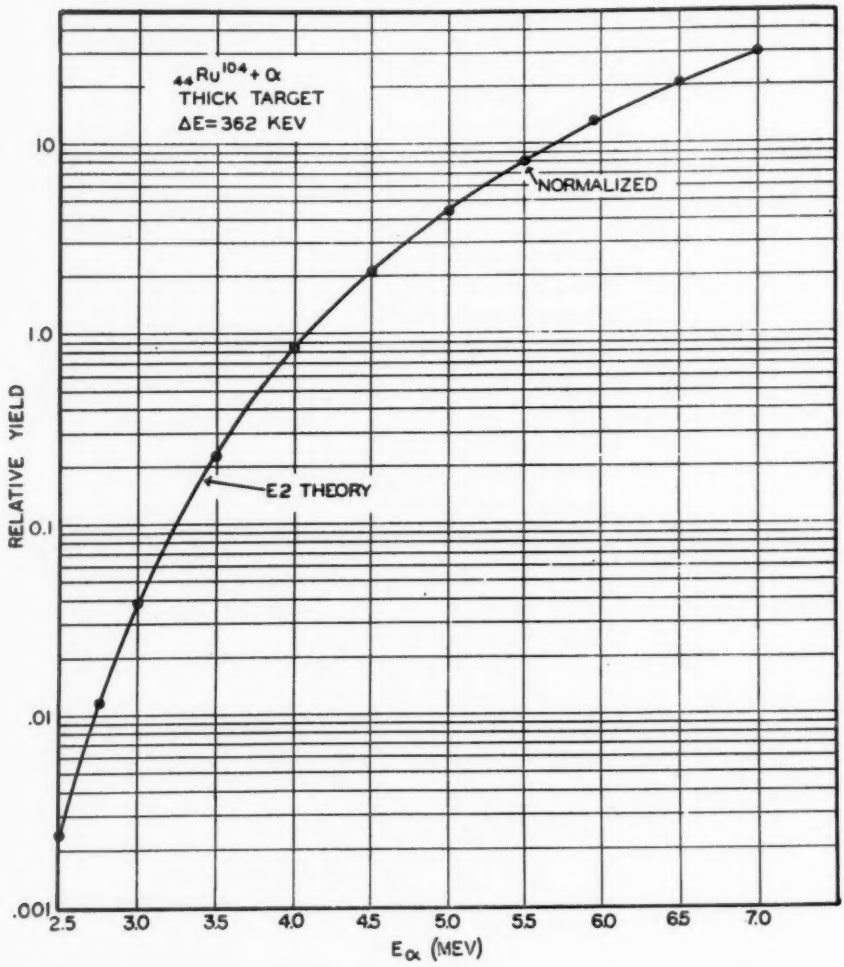

FIG. 8. Thick-target excitation function for 362-kev level in Ru^{104}. Experimental points normalized at 5.5 Mev. Theoretical curve computed from semi-classical theory and using the simplified thick-target treatment outlined under Targets above. Note agreement over a factor of 20,000 in yield. [Taken from Temmer & Heydenburg (105)].

The rate of change with bombarding energy, of the Coulomb excitation cross section (or thick-target yield) depends strongly on the excitation energy ΔE and makes it possible to decide, from a rapid examination of relative gamma-ray (or conversion electron) yields at widely differing bombarding energies, whether a given radiation corresponds to a transition of the same or of higher energy [e.g., Fig. 7 of reference (58)]. The higher the energy of a transition, the steeper its excitation curve; thus for instance, if two gamma-rays maintain the ratio of their yields constant over a wide energy interval, one may conclude that both originate from the same excited state. The usefulness of such considerations in the interpretation of complex decay schemes is evident.

Excitation probability.—Aside from all the extraneous factors affecting the excitation cross section, the intrinsic nuclear quantity of interest concerning the various observed transitions is the reduced transition probability $B(E2)$.[14]

We have already mentioned the model-independent relationship which exists between the lifetime measurement and the excitation cross section measurement of an electromagnetic transition. A large transition matrix element entails a short lifetime and a large Coulomb-excitation cross section, and vice versa. Longer lifetimes ($\gtrsim 10^{-9}$ second) are best suited for electronic delayed coincidence measurements, which have provided the bulk of information on these transitions until very recently (8, 125). Shorter lifetimes ($<10^{-8}$ second) are most accessible to Coulomb excitation measurements, as well as to resonance fluorescence determinations (53, 54). There fortunately exists a region of overlap in the rare earth region and beyond (29, 93, 97), where a number of transitions have been studied in both ways. Within the rather wide limits of experimental uncertainty (of the order of 20 per cent; see below for the reasons) agreement of lifetime and Coulomb-excitation results is found to be good (29, 93, 97). This is of course an essential requirement if Coulomb excitation is to be a trustworthy tool for the measurement of B.

We already mentioned the other possibility for checking Coulomb-excitation B-values for those few nuclei for which resonance fluorescence experiments involving pure $E2$ transitions can be carried out.[15] Comparison can be made for Ge^{72} (53, 95), Ge^{74} (53, 95), Fe^{56} (95, 126), Hg^{198} (104a, 127), and Hg^{202} (104a, 128). There is quite satisfactory agreement between the values obtained by these vastly different methods. There seems to be no systematic departure between the two methods at present and only further fluorescence measurements will show whether definite departures do exist

[14] We shall limit ourselves to the case of $E2$ excitation unless explicitly stated otherwise. We further absorb the factor $1/e^2$ into the quantity B, and can thus express it in units of 10^{-48} cm.^{4} (or square barns) in keeping with the practice of expressing electric quadrupole moments in 10^{-24} cm.^{2} (or barns).

[15] We omit the cases of $M1$–$E2$ mixtures because of the additional uncertainties existing there.

between the transition probabilities determined in these two ways. It should be kept in mind that fluorescence experiments as carried out in practice also involve a certain number of assumptions which need independent check.

Finally, some very preliminary coincidence experiments in the region of $10^{-10}-10^{-11}$ second (129) give promise of some additional checks on Coulomb lifetimes in the future, especially in the medium weight nuclei (see Section III C below).

It should be kept in mind that experimentally observed lifetimes τ_{obs}[16] (especially for low-lying states in medium and high Z nuclei) are shorter than the true gamma-ray lifetimes τ_γ [e.g. see reference (8)] because of the additive contribution from internal conversion; if α_t is the total internal conversion coefficient,

$$\tau_\gamma = (1 + \alpha_t)\tau_{obs} \qquad 13.$$

Similarly, if the Coulomb excitation cross section is measured by gamma-ray detection,

$$B(E2) = (1 + \alpha_t)B_{obs}. \qquad 14.$$

where B_{obs} is the (smaller) quantity derived from the observed cross section through equation 2. We see that observed lifetimes τ_{obs} and observed transition probabilities B_{obs} differ by a factor $(1+\alpha_t)^2$, so that lack of precise knowledge concerning internal conversion coefficients (in all electronic shells) often presents a serious obstacle to decisive comparisons of transition probabilities derived by these different methods. Furthermore, as we mentioned earlier, many $E2$-excited transitions decay by a mixture of $M1$ and $E2$ radiation. In such cases $B(E2)$ of course represents only the $E2$ portion of the transition probability; to relate this quantity to the "observed" lifetime τ_{obs} we must have knowledge of the mixture ratio δ which is usually defined as

$$\delta^2 = \frac{\text{Int. } (E2)}{\text{Int. } (M1)}. \qquad 15.$$

Information concerning δ can be obtained in several ways (80, 82, 86, 90, 94, 103, 109) which will be discussed shortly.

Obvious corrections to B_{obs} must always be made (*a*) if higher states than the one under study are excited and decay by cascade via the latter, or (*b*) if there is more than one way for the state of interest to return to the ground state (branching). In case (*a*) the correction tends to reduce B, while (*b*) requires the summation of all intensity coming from the state in question and raises B. Coincidence measurements may prove very helpful in disentangling such situations (100, 101, 103).

Choice of bombarding particle.—We are of course always interested in maximizing the yield of Coulomb excitation for a given transition of reduced transition probability B. We shall limit ourselves for the present to purely

[16] τ shall always refer to the mean life, related to the transition probability $P = 1/\tau$ sec^{-1}.

kinematic considerations, relegating until later the more indirect factors affecting the success of a given experiment. A glance at equation 2 shows that regardless of projectile, the higher the bombarding energy, the higher the excited states we can reach, or the higher the atomic number of the nucleus we can excite effectively. However, since one is usually limited by the energy available from a given accelerator, it is worth comparing the relative effectiveness of, say, protons and alpha particles at the same machine energy. Bjerregaard & Huus (28) have presented a graph of this type (including deuterons) which is useful for thin target work, such as is necessary for internal conversion detection. Since most Coulomb excitation experiments use gamma-ray detection and thick targets, we think it worthwhile to present in Table III the approximate relative effectiveness for the excitation of 100-kev and 500-kev transitions, for two bombarding energies (3 Mev and 6 Mev), two kinds of particles (p and α), and thick targets of

TABLE III

RELATIVE THICK-TARGET COULOMB EXCITATION YIELDS FOR VARIOUS PARTICLES AND TARGETS, AND FIXED REDUCED TRANSITION PROBABILITY $B(E2)$

ΔE (kev)	Z_2	$E=3$ Mev		$E=6$ Mev	
		H^+	He^+	H^+	He^+‡
100	30	6.12 (0)*	1.78 (0)	3.52 (1)	1.54 (1)
	60	*1.00*†	1.30 (−1)	6.17 (0)	2.35 (0)
	90	2.88 (−1)	1.87 (−2)	2.20 (0)	6.23 (−1)
500	30	2.59 (0)	1.79 (−2)	3.25 (1)	4.80 (0)
	60	1.22 (−1)	7.59 (−6)	3.84 (0)	1.37 (−1)
	90	1.02 (−2)	6.11 (−9)	9.66 (−1)	6.96 (−3)

* Numbers in parentheses are power-of-ten multipliers.

† All entries arbitrarily normalized to *1.00* for 3-Mev protons, $Z_2=60$, and $\Delta E=100$ kev.

‡ This column also for He^{++} ions at V = 3 Mev.

$Z_2=30$, 60, and 90. We assume the same reduced transition probability B throughout, as well as the simplified thick-target treatment discussed earlier (for pure elemental targets), and normalize to *1.00* for proton excitation at $\Delta E=100$ kev, $E=3$ Mev and $Z_2=60$. He^{++} values for 3 Mv. accelerating voltage can of course be read from the He^+ column at $E=6$ Mev.

Now in addition to the more obvious factors occurring explicitly in equation 2 there are a number of other considerations which enter when making a choice as to bombarding particle for a transition of a given strength B. We

shall assume that it is desirable to obtain the maximum possible "signal-to-noise" ratio in a Coulomb excitation experiment and limit ourselves to intrinsic sources of "noise" (i.e., noise coming from the target atoms of interest). For gamma-ray detection, the following sources of noise exist: (*a*) radiation from compound nuclear reactions, either leading to the same gamma-ray transition by inelastic scattering, or to other gamma rays causing undesirable background in the detector (58, 60); (*b*) bremsstrahlung from the bombarding particle (55, 93, 130, 131) giving rise to a continuum of gamma radiation; (*c*) characteristic x-radiation emitted from the target (6, 77, 130, 132). It turns out that all these undesirable effects are minimized by using heavier projectiles [to date alpha particles have been the heaviest particles used to any great extent; see however (27a)].

For radiation (*a*), it has been shown that the higher Coulomb barrier for higher Z_1 projectiles insures an adequate range of bombarding energies in which Coulomb excitation is predominant, even for rather low Z_2 targets (58, 105), so that meaningful transition probabilities can be extracted from the measured cross section values. Bremsstrahlung (*b*) is a component which must always be subtracted when using protons (60, 93); heavier particles generally contribute negligible quantities of bremsstrahlung (55, 130) Finally, the characteristic *K* and *L* x-rays emitted under bombardment must always be reckoned with. Because of the dependence of the yield of this radiation on Z_1^2/A_1^4 (29, 130), heavier particles contribute much less (77, 130); for instance, at a given bombarding energy, alpha particles produce about 1/64th the *K* x-rays of protons. It should be remembered, however, that an appreciable fraction of this x-radiation in the heavier elements is often projectile-independent, because it results from the filling of an electronic shell vacated by the internal conversion of a Coulomb-excited gamma transition. For instance, in the case of Ta^{181} excited by alpha particles, nearly all of the Ta *K* x-rays are accounted for in this way [see Fig. 1 of reference (77)]. A judicious choice of absorbers will usually reduce this x-radiation to a tolerable level regardless of bombarding particle.

If we are interested in the detection of internal conversion electrons, the only disturbing radiation emanating from the target are the δ-rays or target electrons (29, 130), which are the counterpart of the target x-rays discussed earlier, and make low-energy electron detection (i.e., electrons coming from the conversion of low-energy transitions in the *K*-shell) difficult. Once again, this problem is minimized when using heavier bombarding particles (29, 109). Figure 4(a) above illustrates the *K*, *L* and *M* conversion lines from the famous 137-kev transition in Ta^{181} as observed under alpha-particle bombardment (109).

Angular distribution of gamma radiation.—An important survey of the angular distribution of gamma radiation following the Coulomb excitation, by protons, of medium-heavy and heavy elements was carried out (94) using thick targets. Although the results showed apparently serious discrepancies with the then existing theory, (see Coulomb Excitation Theory), they demon-

strated the great usefulness of such experiments in determining spins of excited states, as well as of $E2/M1$ mixture ratios in the decay of odd-A nuclei. By gathering enough information on $0^+ - 2^+$ transitions in many even-even nuclei, McGowan & Stelson were thus able to determine the empirical systematic behavior of the coefficients a_2 and a_4 of equation 9 (see theoretical section above) as a function of ξ, for thick targets. The thick targets gave rise to a slight Z_2 dependence over and above the ξ dependence. Using these empirical curves, one is now able to determine mixtures of radiation from Coulomb excited states in neighboring odd-mass nuclei, independently of either nuclear or Coulomb excitation theory. The mixture ratios δ defined in equation 15 will usually be double-valued because of the nature of the anisotropy *versus* mixture relation. The case of ground-state spin $I_0 = \frac{1}{2}$ is particularly unambiguous as to spin assignment, since any $E2$-excited state must have either spin 3/2 or 5/2, and the mere presence of an $M1$-$E2$ mixture establishes the spin as 3/2. Pure $E2$ gamma radiation always leads to a positive anisotropy[7] (i.e., more radiation parallel to the beam than at right angles), while mixed $M1$-$E2$ radiation may give rise to either positive or negative anisotropies, depending upon the relative amplitudes and phases of the $M1$ and $E2$ matrix elements (see references 64 and 66). In many cases the spin assignments to excited states may be ambiguous for this reason. However, as was pointed out above, polarization measurements can and have been used successfully to select a unique value for the mixture ratio δ (75a).

C. Discussion and Interpretation of Coulomb Excitation Results

Vital statistics.—Nearly 200 transitions have been excited in over 120 stable nuclei to date, the great majority of them by alpha-particle bombardment.[17] About one third of these states were not known from previous nuclear spectroscopy for reasons varying from the purely technical impossibility of reaching some states from neighboring isobars, to the conspicuous absence of transitions due to the operation of some selection rules of beta or gamma decay. Comparatively few nuclei do not show any excited states, the reason usually being kinematic in the sense discussed under *Excitation probability*; some instances of non-observation of well-known levels have also been found (98), their absence being due to the "wrong" multipolarity (other than $E2$) of the transitions involved.

Medium-weight nuclei of the even-even type ($A \lesssim 150$).—All stable nuclei of this type having first-excited states below about 1 Mev have been excited, giving rise to one and only one gamma-ray (about 70 cases). The well-known, systematic behavior as to position, spin, and parity of these states (123, 124) has been confirmed and extended to their lifetimes by Coulomb

[17] For a compilation of these transitions, as well as an extensive discussion by the Copenhagen group of Coulomb excitation in theory and experiment, the reader is referred to reference (56).

excitation (93, 97, 105). For some time so-called "fast" $E2$ transitions were known to exist in the even-even rare earth nuclei where they were accessible to electronic measurement (125), "fast" being understood in the sense that their reduced transition probabilities $B(E2)$ were about two orders of magnitude larger than would be estimated for the transition of a single proton.[18] Coulomb excitation has shown that essentially all even-even transitions in the medium-heavy (non-magic) nuclei[19] (roughly from Ti to Te) are from 10 to 30 times faster than a single-particle transition (60, 92, 97, 105) and must therefore involve some degree of collective nuclear participation (135, 136), although probably not of the simple variety encountered in strongly deformed nuclei because their equilibrium deformations are not large enough (see below). In Figure 9 we summarize some results on medium-weight even-even nuclei (97); whereas the exact meaning of the ordinates β_E and β_B is doubtful for these lighter nuclei, these parameters are essentially proportional to $(\Delta E)^{-1}$ and $B(E2)$, respectively (see Strongly deformed nuclei below) and illustrate the degree of correlation which exists between these two quantities as well as the influence of closed shells. This type of information is of a kind which became available only with the advent of Coulomb excitation, and is bound to add much to our knowledge about nuclei in this region.

Medium-weight nuclei of the odd-A type ($A \lesssim 150$).—As might be expected, the excitation results on medium-weight odd-A nuclei inherently do not lend themselves to a general systematic description (see however, Strongly deformed nuclei below). Since their levels almost invariably lie lower than in neighboring even-even nuclei, they are more easily excited and studied (58, 105, 100). With the possible exception of the spin $\frac{1}{2}$ nuclei Ag^{107}, Ag^{109}, and Rh^{103} (83, 94), no clearly discernible trends in $E2$-excited states are evident to us for odd nuclei between fluorine and barium. The number of levels excited in each nucleus varies from one to seven; transition probabilities fluctuate greatly, from less than to many times greater than single-particle strength, without apparent regard as to the nature of the odd particle or the location of the nucleus on the nuclear chart.[20] Although a considerable body of empirical knowledge concerning these nuclei already exists, it is clear that more theoretical progress will have to be made with their even-even neighbors before the more intricate problem of the individual odd-A nuclei, with their wide, individual variations in coupling schemes, has any hope of being tackled successfully. For an interesting summary plot of the reduced life-

[18] The single-proton transition probability $B_{sp}(E2)$, although not expected to be a realistic measure of transition rate, is nevertheless a useful unit of measurement ["Weisskopf" unit (133, 134)] for the comparison of different transitions. A rough estimate of this unit for a $0^+ - 2^+$ (upward) transition, using a nuclear radius $R_0 = 1.2 \times 10^{-13} A^{1/3}$cm., is $B_{sp}(E2) \cong 3 \times 10^{-63} A^{4/3}$ cm.^{4}

[19] See reference (137) for a good empirical definition of "medium-weight" nuclei.

[20] This is not to imply however, that there are no definite trends in odd-A nuclear properties studied by other means; see (137).

times [as defined in (8)] of all medium-weight nuclei, as well as a more thorough discussion of the properties of these nuclei, we refer to a companion article in this volume (137).

Strongly deformed nuclei of the even-even type.—There is considerable evidence that certain nuclei, located far enough from closed shells, depart so strongly from the spherical shape in their ground states that the so-called

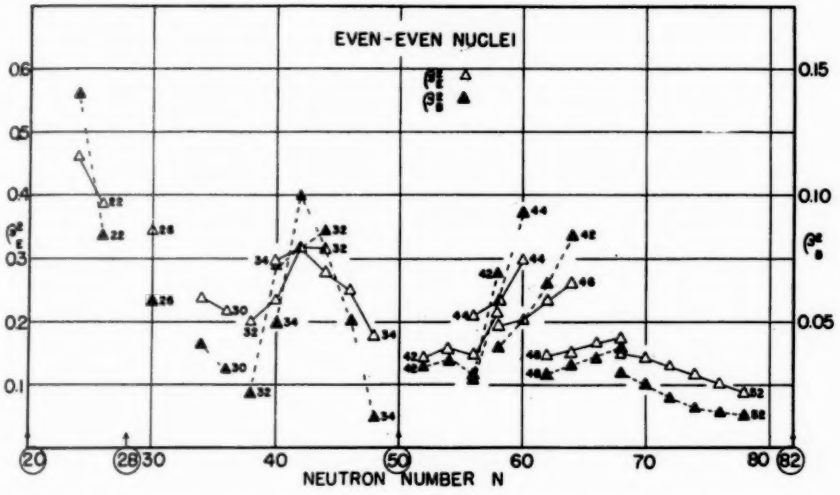

FIG. 9. Summary of some Coulomb excitation results for medium-heavy even-even nuclei. Empty symbols represent "deformations" obtained from position of first-excited 2^+ states and have ordinates inversely proportional to ΔE; full symbols represent "deformations" obtained from reduced quadrupole transition probabilities to the 2^+ states, and have ordinates proportional to $B(E2)$. Atomic numbers of various elements are given in the diagram. Note factor of four between left hand and right hand scales; also the remarkable correlation between β_B and β_E. For definition of deformations, see text. [A revised plot taken from Temmer & Heydenburg (97).]

"strong coupling" approximation of the unified model (52) which presupposes the existence of equilibrium deformations large compared to the inherent quantum fluctuations in this deformation, is applicable, predicting the existence of simple, rotational energy states (bands) as well as enhanced $E2$ transition rates within these bands. These conditions seem to be satisfied in the rare earth region, and again in the actinide and transuranic element region; more specifically, nuclei having neutron number $N \geqq 90$ and $N \leqq 112$, and again $N \geqq 136$. We refer the reader to a number of other sources on the subject of the unified nuclear model and rotational states for a more detailed discussion (52, 138 to 141). Because of the characteristic spin sequence of these states in even-even nuclei (0^+, 2^+, 4^+, 6^+ . . .) Coulomb excitation cannot contribute spacings to be compared with the theoretical predictions

(other than the first). It has however much to offer by way of the systematic variation of the transition rate to the 2^+ state from nucleus to nucleus. The transition probability for the $0^+ - 2^+$ transition in an even-even nucleus is given by

$$B(E2) = \frac{5}{16\pi} Q_0^2. \qquad 16.$$

Q_o is the intrinsic nuclear quadrupole moment which plays such a prominent role in this model, and is approximately given by

$$Q_0 \cong \frac{3}{\sqrt{5\pi}} ZR_0^2\beta = 0.011\ ZA^{2/3}\beta \text{ barns} \qquad 17.$$

for a spheroidal shape, R_0 being the (average) nuclear radius, and β a distortion parameter which equals $1.06\Delta R/R_0$, where ΔR is the difference between semi-major and semi-minor axes. Q_0 is defined with respect to a body-fixed system of axes; it is meaningful for all strongly deformed nuclei regardless of spin, and hence for even-even nuclei as well. Such values as exist for spectroscopic quadrupole moments Q_{hfs} in deformed odd-A nuclei generally agree with those obtained from transition rates. It must be remembered that Q_{hfs} is measured with respect to a laboratory-fixed frame of reference, and is related to the (larger) intrinsic Q_0 by the expression

$$Q_{hfs} = \frac{I_0(2I_0 - 1)}{(I_0 + 1)(2I_0 + 3)} Q_0. \qquad 18.$$

Figure 10 gives a plot summarizing much of the information obtained from both lifetime and Coulomb-excitation measurements for deformed even-even nuclei. The values of β_B^2 derived from lifetimes are taken from the compilation of Sunyar (125), while those for Coulomb excitation are obtained from equations 16 and 17 and the results of Temmer & Heydenburg (97, 98), and are directly proportional to $B(E2)$ [$\beta_B^2 = 8.48 \times 10^4 B(E2)/Z^2A^{4/3}$; B($E2$) in units of square barns]. Similar plots can be found in (8) and (13). Values for β_E^2 are obtained from the relation $\beta_E^2 = 2.43 \times 10^5/A^{5/3}\Delta E(\text{kev})$, and hence are inversely proportional to the energy ΔE of the 2^+ states. As far as the absolute magnitudes of the level spacings are concerned, which depend directly on the effective moment of inertia of "what rotates," a long-standing discrepancy between deformation deduced from transition strengths (β_B) and those inferred from level positions (β_E) has recently been resolved (142, 143) so that the factor shown between left hand and right hand ordinates in Figure 10 has now been explained. Only β_B should be considered meaningful in an absolute sense in these plots.

Strongly deformed nuclei of the odd-A type.—Because of the characteristic spin sequence in rotational bands of odd-A nuclei (I_0, I_0+1, I_0+2, I_0+3, . . . , all of the same parity as ground state), it is possible to excite the first two excited states by $E2$ excitation and compare the ratio of their spacings with the predicted values. In fact, Coulomb excitation has contributed

most of the original information in the rare-earth region (29, 81, 98) and in all cases of strong deformation ($153 \leqq A \leqq 187$, about 15 nuclei) the observed ratios agree with the predicted ones within the experimental error (about one per cent) regardless of ground state spin I_0 (29, 98). In one instance (Hf^{177}) it was possible from the spacings to correctly predict (98) the

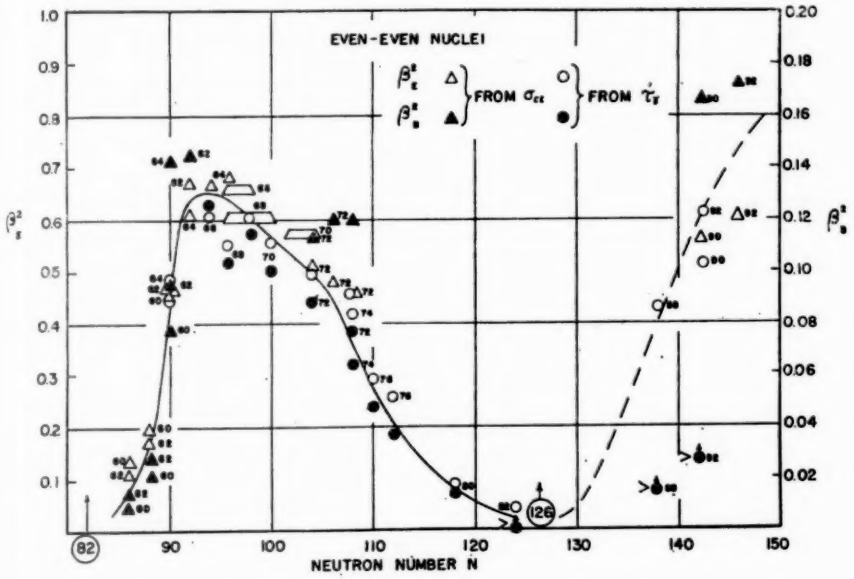

FIG. 10. Summary of some Coulomb excitation results for deformed, even-even rare earth nuclei. Definitions similar to those given in the caption of Figure 9. Triangles refer to Coulomb excitation as before; circles refer to results from lifetime and resonance fluorescence measurements as summarized by Sunyar (125). Note factor of five between left hand and right hand scales. For definition of deformations, see text. Note large gap in deformations upon passing from $N=88$ to $N=90$. [A revised plot taken from Temmer & Heydenburg (97).]

then unknown ground-state spin (144). In the region of the transuranic elements, considerable information on odd-A nuclei exists from alpha-decay studies (141); it was also possible to obtain interesting Coulomb excitation data in three instances (106). We summarize results on odd-A level spacings in Table IV.

Expressions similar to equation 16 above exist for the transitions $I_0 \rightarrow I_0 + 1$ and $I_0 \rightarrow I_0 + 2$ in strongly deformed odd-A nuclei:

$$I_0 \rightarrow I_0 + 1: B(E2) = \frac{15}{16\pi} Q_0^2 \frac{I_0}{(I_0 + 1)(I_0 + 2)} \qquad 19.$$

$$I_0 \rightarrow I_0 + 2\colon B(E2) = \frac{15}{16\pi} Q_0^2 \frac{2}{(2I_0+3)(I_0+2)}. \qquad 20.$$

The ratio of equations 19 and 20, which is independent of Q_0, has been tested and confirmed in a number of experiments on odd-A nuclei (6, 29, 98, 103). It must be remembered that both quantities B as observed must be corrected for cascade and branching in the manner discussed under Excitation proba-

TABLE IV

ROTATIONAL STATES IN STRONGLY DEFORMED ODD-A NUCLEI REACHED BY COULOMB EXCITATION

Nucleus	I_0*	E_{I_0+1}(kev)	E_{I_0+2}(kev)	ρ_{exp}†	ρ_{theor}‡	References
$_{63}Eu^{153}$	5/2	82	187	2.28±0.04	2.29	(98)
$_{64}Gd^{155}$	3/2	60	146	2.43±0.07	2.40	(111), (98)
$_{64}Gd^{157}$	3/2	55	132	2.42±0.07	2.40	(111), (98)
$_{65}Tb^{159}$	3/2	57	136	2.39±0.05	2.40	(29), (98)
$_{66}Dy^{161,163}$§	(7/2)	76	166	2.18±0.04	2.22	(98)
$_{67}Ho^{165}$	7/2	94	206	2.19±0.04	2.22	(98)
$_{68}Er^{167}$§	7/2	79	172	2.18±0.04	2.22	(98)
$_{70}Yb^{173}$§	5/2	78	180	2.31±0.04	2.29	(98)
$_{71}Lu^{175}$	7/2	114	250	2.19±0.04	2.22	(98), (91)
$_{72}Hf^{177}$	7/2	112	250	2.23±0.04	2.22	(98), (144)
$_{72}Hf^{179}$	9/2	119	260	2.18±0.04	2.18	(98), (144)
$_{73}Ta^{181}$	7/2	136	303	2.23±0.04	2.22	(6), (80), (82)
$_{75}Re^{185}$	5/2	126	286	2.27±0.05	2.29	(91), (101a), (104)
$_{75}Re^{187}$	5/2	135	303	2.25±0.05	2.29	(91), (101a), (104)
$_{92}U^{233}$	5/2	40.5	92.1	2.27±0.02	2.29	(106)
$_{92}U^{235}$	7/2	46.5	103.8	2.23±0.02	2.22	(106)
$_{93}Np^{237}$	5/2	33.5	76.4	2.28±0.02	2.29	(106)

* $I_o = \frac{1}{2}$ nuclei (unconventional spectra) excluded: $_{69}Tm^{169}$, $_{70}Yb^{171}$, $_{74}W^{183}$, $_{94}Pu^{239}$

† $\rho_{exp} = E_{I_o+2}/E_{I_o+1}$.

‡ $\rho_{theor} = (2I_o+3)/(I_o+1)$.

§ Conclusions tentative, based on unseparated targets.

bility above. Note also that for identical intrinsic quadrupole moments, the quantities $B(E2)$ leading to the first-excited states of even-even nuclei are about half the $B(E2)$ for odd nuclei [see equations 16 and 19]. No clearcut evidence has been uncovered for intrinsic differences in deformation between even and odd nuclei (29, 98, 103) as derived from transition probabilities, whereas, the "moments of inertia" of odd nuclei seems to be slightly larger (98).

For the reduced magnetic dipole transition probability $B(M1)$ between states differing in spin by unity the model gives

$$I_o + 1 \rightarrow I_o\colon\ B(M1) = \frac{3}{4\pi}\mu_N^2 (g_\Omega - g_R)^2 \frac{I_0^2(2I_0+1)}{(I_0+1)(2I_0+3)} \qquad 21.$$

where μ_n stands for the nuclear magneton, and g_Ω and g_R are the intrinsic and collective g-factors, respectively. The former is associated with the motion of the odd particle in the deformed potential (145), while the latter embodies the gyromagnetic contribution of the aggregate of particles in the deformed nucleus. The quantity $B(M1)$ (equation 21) can be obtained from the Coulomb excitation of deformed odd-A nuclei by determining the $M1$-$E2$ amplitude mixture ratio for the rotational transitions in a given band (*a*) from the internal conversion coefficients or ratios in different shells (29, 109); (*b*) from gamma-ray angular distribution measurements (see General Features of Coulomb Excitation Experiments above); (*c*) by determining the branching ratio between the cross-over and cascade gamma transition from the second rotational state (103). Methods (*a*) and (*b*) are model-independent, while (*c*) depends on the correctness of relations 19 and 20 which are fairly well established. A useful relation between the mixture ratio δ and the quantities involved in $M1$ and $E2$ transitions can be derived from equations 4, 19, and 21:

$$I_o + 1 \rightarrow I_o\colon\ \delta = \frac{0.933\,\Delta E\, Q_o}{(g_\Omega - g_R)\sqrt{I_0(I_0+2)}} \qquad 22.$$

where ΔE is the level splitting in Mev, and Q_o is the intrinsic quadrupole moment in barns; δ may be positive or negative. Since another relation exists, for strongly deformed nuclei, between g_Ω and g_R in the expression for the static ground-state magnetic moment μ, namely

$$\mu = \frac{I_o}{I_o+1}(g_\Omega I_o + g_R), \qquad 23.$$

it is possible to obtain these quantities separately. Table V shows a summary of these determinations. g_Ω can be computed from the ground-state configurations and appropriate spheroidal-well wave functions (145). Although g_R is expected to be near $Z/A \sim 0.4$, significant deviations may be discerned to either side of that value. The excess found in some odd-Z nuclei (and the decrease in odd-N nuclei) may well be related to the comparatively large share of the total angular momentum and moment of inertia contributed by the odd particle (146).

Finally we may ask the question: are there any other states in strongly deformed nuclei which should be accessible by Coulomb excitation? In addition to the rotational states discussed above, the theory predicts vibrational states (52, 138, 140) at considerably higher energies, (the first between 1 and 2 Mev) of which one should be reached via enhanced $E2$ transition in even-even nuclei, and possibly several in odd-A nuclei. No definite evidence for such states exists at present. A further possibility exists in so-called rotational perturbations (147), where certain low-lying intrinsic particle states (espe-

TABLE V

COULOMB EXCITATION RESULTS ON INTRINSIC AND COLLECTIVE g-FACTORS FOR STRONGLY DEFORMED ODD-A NUCLEI

Nucleus	I_0	μ_0(n.m.)	ΔE (kev)	Q_0 (barns)	δ^2*	Method†	g_Ω‡	g_R‡	Ref.
$_{63}Eu^{153}$	5/2	+1.6	105§	7.4	0.38	(b)	0.72	0.45	(103)
$_{64}Gd^{155}$	3/2	−0.30	60‖	7.6	0.05	(a)	−0.53	0.28	(111), (148)
$_{64}Gd^{157}$	3/2	−0.37	55‖	8.1	0.04	(a)	−0.61	0.27	(111), (148)
$_{65}Tb^{159}$	3/2	+1.5	79§	8.7	0.013	(b)	1.70	∽0**	(103)
$_{67}Ho^{165}$	7/2	+3.3	112§	7.3	0.039	(b)	1.10	0.39	(103)
$_{71}Lu^{175}$	7/2	+2.9	136§	8.0	0.13	(b)	0.92	0.50	(103)
$_{72}Hf^{177}$	7/2	+0.61	138§	8.0	1.64	(b)	0.21	0.06	(103), (144)
$_{72}Hf^{179}$	9/2	−0.47	141§	6.9	0.15	(b)	−0.04	−0.40	(103), (144)
$_{73}Ta^{181}$	7/2	+2.1	137‖	7.7	0.25	(a)	0.70	0.25	(93)

* Mixture ratio defined in equation (15). Values derived from either cascade transition predicted to be equal within 10 per cent.

† (*a*) from internal conversion interpolation: (*b*) from crossover-cascade branching ratio.

‡ Because of ambiguity in sign (cf. equations 21 and 22) there exist alternate pairs of solutions in all cases. We have arbitrarily selected those yielding the smaller value of g_R.

§ $I_0+2\rightarrow I_0+1$ transition.

‖ $I_0+1\rightarrow I_0$ transition.

** Value extremely sensitive to δ^2.

cially in odd-A nuclei) may be excited by the collective rotation, i.e., the rotational motion is no longer strictly separable from the intrinsic degrees of freedom. This coupling results in larger than "normal" (single-particle) $E2$ transition probabilities and should be favored in Coulomb excitation.

IV. SUMMARY

We have chosen to emphasize in this article those aspects of Coulomb excitation bearing on its usefulness and scope as a universal tool for the exploration of nuclear properties in all regions of the nuclear chart. While we have discussed some of the inescapable conclusions to be drawn from these experiments in terms of a particularly simple limiting case of the unified model (the strong-coupling approximation), we have by no means done justice to all theoretical aspects of this rapidly growing subject; we refer the reader to a number of important sources (52, 56, 142) for discussions more specifically concerned with the unified model and its relation to Coulomb excitation.

We have shown that it is relatively straightforward to determine energies, spins, parities, and lifetimes of excited states of stable nuclei, as well as the

character and mixture of the gamma radiation connecting them with their ground states. The method of Coulomb excitation (with heavy particles) is without doubt one of the best-understood tools for the study of these low-lying states. The uniformity of this method as we move from nucleus to nucleus is undoubtedly its chief virtue; it is small wonder that most of those nuclear properties which do not change abruptly between nuclei have been especially highlighted. The privileged position occupied by the electric quadrupole component of the electromagnetic field in the Coulomb-excitation process on the one hand, and by the electric quadrupole matrix in the collective behavior of nuclear matter on the other hand, has provided a natural bridge between experiment and theory, with great benefit to both. In fact, we feel that in regions where the existence of rotational states is well established (rare earths, actinide, and transuranic elements) Coulomb excitation represents the best available tool for the measurement of static electric quadrupole moments, being altogether independent of the details of electronic structure which are, at best, poorly known.

We should, however, not lose sight of the fact that Coulomb excitation yields most facts concerning nuclear excited states independently of any nuclear model.

We have placed less emphasis on the field of high-energy electron excitation, mainly because, from an experimental standpoint, it is still in its infancy; we may however reasonably expect much additional and novel information from that quarter in the not too distant future.

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ISOTOPE SHIFT IN ATOMIC SPECTRA[1]

By J. E. Mack

Department of Physics, University of Wisconsin, Madison, Wisconsin

AND

Hack Arroe

Department of Physics, Montana State College, Bozeman, Montana

1. Introduction

In the spectrum of an atomic system consisting of a nucleus with proton number (atomic number) Z and neutron number N, in the nuclear ground state, surrounded by z electrons in states characterized by electron quantum numbers a, the transition energies ΔE or the wave numbers $\sigma = \Delta E/hc$ of the lines depend approximately only upon Z, z, and a; but when studied more closely they are found to depend also slightly upon N. The N-dependence, or so-called isotope shift, is the subject of this review.

If the (Z,N) nucleus has a non-zero angular momentum, the levels and lines in general exhibit hyperfine-structure splitting. In isotope shift studies the hyperfine structure itself need not be treated: each hyperfine-structure multiplet may be treated as a single level or line at the centroid (except that those few cases where fine and hyperfine structure separations are of the same order of magnitude need further, straightforward treatment).

The most comprehensive compilation of isotope shift in atomic spectra is that of Brix & Kopfermann in *Landolt-Börnstein* (1), which covers the literature to 1950. The subject is discussed extensively in the revised edition of Kopfermann's book (2) and several general theoretical reviews have appeared (3, 4, 5).

One respect in which isotope shift studies differ markedly from most other spectroscopic studies is that the small energy differences under investigation occur between level pairs in different atoms, and any common reference point is purely arbitrary. In isotope shift studies involving more than one line, a convenient custom is to consider the spectra under consideration to be arbitrarily matched at a series limit appropriate to the lines under study, the limit usually, but not always, being the ground level of the next higher stage of ionization. The usual (but not universal) convention with respect to the sign of the shift, which is the one used in this review, is to consider the direction of the normal mass shift (discussed below) positive; thus the level shift is called positive when the level belonging to the heavier of two isotopes is farther below the reference series limit than the corresponding level of the lighter isotope, and the line shift is called positive when, between corresponding lines, that belonging to the heavier isotope has the higher wave number.

The treatment of isotope shift can be carried out under two headings: mass effect and field effect. The former depends only upon the nuclear mass and the electron wave functions, while the latter depends upon details of the

[1] The survey of literature pertaining to this review was completed in July, 1956.

nuclear structure. The former tends to decrease and the latter to increase with increasing Z, and while the observations cannot be separated into two categories quite as consistently as the theory, it is roughly true to say that the effect in the light elements is predominantly a mass effect and that in the heavy elements a field effect, while in the intermediate region ($Z \sim 20$ to 40) the shift is very small.

The general features of the isotope shift observed in atomic spectra can be crudely illustrated schematically (Fig. 1). Figure 1a shows a term shrink-

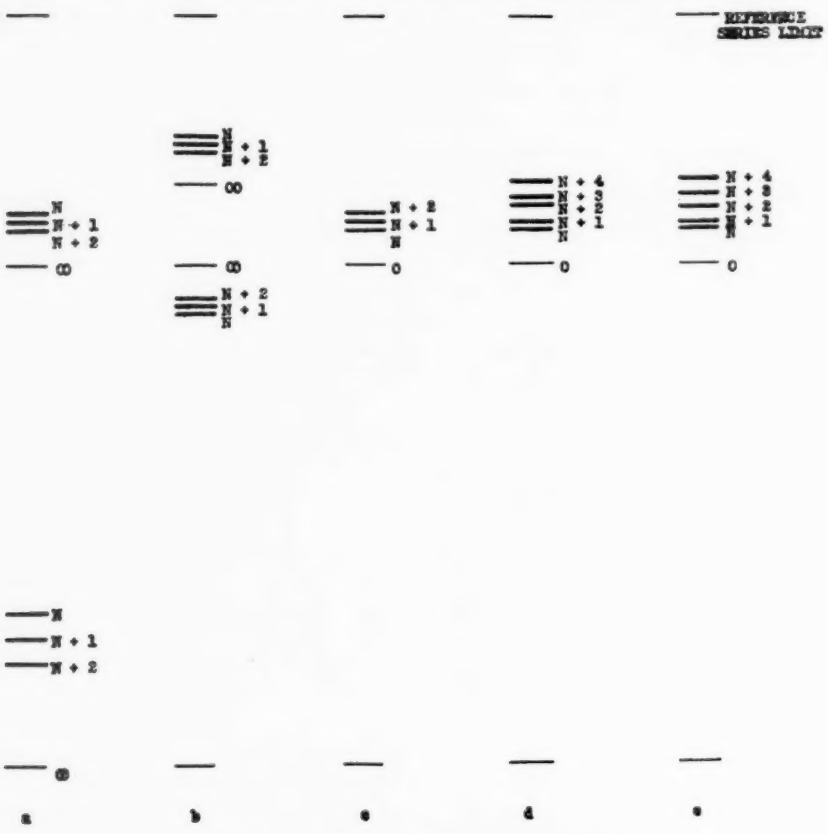

FIG. 1. General features of atomic isotope shifts.

age that occurs in all spectra, and is the only shift in the spectra in the hydrogen-like isoelectronic sequence: the shift (measured from the value postulated for an infinitely massive nucleus) tends to reduce the term differences and is strictly proportional to the term value and to the reciprocal of the mass of the atom or ion; thus the separation between adjacent mass numbers is approximately inversely proportional to the square of the average mass. This universal term shrinkage is completely accounted for as the normal mass effect, discussed below. For simplicity it is not shown in the

other parts of Fig. 1, all of which show selective shifts, i.e., shifts occurring only in certain levels.

In its simplest manifestation (Fig. 1b), from $Z=2$ through about the first third of the atomic table, the selective shift of a level may be of either sign, depending on the electron quantum numbers; and the magnitude of the shift per unit mass change for those levels that are most strongly affected in any spectrum is somewhat smaller than the normal mass effect and varies from element to element in roughly the same way, i.e., inversely as the mass squared, or approximately linearly in the mass when the mass differences are small compared with the mass itself. This shift is identified with the specific mass effect.

All the following phenomena are attributed to the field effect, and the usual hypothetical reference level is not that with infinite mass but that with zero volume (zero mass). About where the specific mass effect becomes inappreciable ($\sim 10^{-3}K$, where K stands for the Kayser, or cm^{-1}) there begins to be noticeable, but only among certain levels (Fig. 1c), an increase in the energy with increasing mass. The energy change depends only upon the configuration, and is greatest in the case of a configuration with a single unbalanced s-electron; but for a $p_{1/2}$-electron it grows to about one-fifth this magnitude in heavy atoms. Notice that the only characteristic distinctions to be observed experimentally between the specific mass effect (Fig. 1b) and the field effect when the latter is approximately linear (Fig. 1c) are that the former may have either sign while the latter is always negative; also, the two effects differ markedly with respect to their dependence upon the electronic quantum numbers.

Figure 1d illustrates odd-even staggering, which usually occurs in the field shift and consists in the relative lowering of a level in an atom with odd neutron number to a position nearer that for the next lighter than for the next heavier atom.

Figure 1e shows the most general situation, which is beginning to be recognized as the usual one in the field-effect region of the atomic table: even for increases of 2 in N (i.e., when N stays odd or stays even) the position of the level is appreciably nonlinear in N.

2. Mass Effect

The mass effect is the manifestation of the fact that the nucleus is not at the center of mass, but in common with the electrons, moves about the centroid of the system. Reference of the nonrelativistic Schrödinger equation to the center of mass leads (6) to a term quadratic in the momenta that can be analyzed into two energy correction terms that would vanish if the nuclear mass were infinite.

One term (normal mass effect) contains only squares of the momenta of single electrons and, exactly as in classical mechanics, leads to a shrinking of the whole spectrum by a factor m_r/m_e or $m_K/(m_e+m_K)$, where m_e is the electron mass, m_K the nuclear mass, and m_r the reduced mass of the system. The other term (specific mass effect) contains only cross products of the momenta of the electrons. In the approximation in which each electron is

supposed to be acted on by an attractive force centered at the nucleus (exchange mass effect) (6) the only electron pairs that participate are those in which the l-values differ by ± 1. The actual force on an electron includes the Coulomb repulsion of every other electron, which is phase-dependent and gives rise to an electron polarization mass effect, small compared with the exchange mass effect, that affects all electron pairs.

The difference in isotope effect between the relativistic and nonrelativistic two-body problems is small (7). Modern work on the former has been done by Breit's group (8, 9) and others (10, 11). (The classification here under mass effect rather than field effect is arbitrary.)

The normal mass effect is generally recognized to be a closed subject. The data for the case $Z=z=1$ are very precise and are susceptible to relatively simple and exact calculation, free from the influence of the specific mass effect; they are among the basic data from which the universal physical constants are determined (12, 13, 14). In the study of other isotope effects, the observed data are ordinarily corrected for the normal mass effect.

The simplest atom that can exhibit a specific mass effect is helium ($Z=2$). The isotope shift in ${}_2$He I has been the object of extensive study in the past decade, both experimental (15 to 18) and theoretical (19, 20).

The experiments of Bradley & Kuhn (17) and those of Fred *et al.* (18) on ${}_2$He I overlap considerably and agree very well in most, but not in all, details. After correction for the normal mass effect the ${}^1P$ levels exhibit a positive, and the ${}^3P$ levels a negative residual, decreasing somewhat more rapidly with approach to the series limit than if they were directly proportional to the term value. This behavior is in qualitative agreement with the Hughes-Eckart theory (6), although the magnitudes are rather greater than predicted from the hydrogenic wave functions used in the original calculation. The ${}^3S$ and ${}^1S$ levels show a positive residual shift about one-fifth as great that can only be accounted for as polarization mass effect; for the $1s$ $2s$ levels see Table I below. The residual for the ${}^3D$ and ${}^1D$ levels is zero within the error of the experiment, except for a phenomenon reminiscent of resonance repulsion, centered at about $1s8d$ ${}^{3,1}D$, just where, in the observed spectrum, there appears to be an actual inversion of the usual singlet-triplet difference. Any attempt to interpret the apparent resonance ought to await a verification of the inversion, based upon intercombinations or long series of both.

The calculations of Bethe (21) on the polarization mass effect for the ground level of ${}_2$He I have been corrected and replaced by the machine results of Wilets & Cherry (22). No isotope shift observation has ever been made on this level. Stone (20) has calculated the ${}_2$He I levels $1s2s$ ${}^3S,{}^1S$ with the self-consistent field wave functions of Huang (23) and corrected Hylleraas-Undheim (24) functions. Another set of calculations (19) based on Huang's functions, yielding smaller values, is probably invalid (25). The data are compared in Table I. In the present state of the study the agreement must be considered very satisfactory.

The spectrum of ${}_3$Li I has been studied in detail by R. H. Hughes (26). Qualitatively somewhat as in ${}_2$He I, it exhibits a negative ${}^2P$ term shift

TABLE I

SPECIFIC MASS SHIFT IN THE LEVELS $_2$He I $1s2s$ $^3S_1$, $^1S_0$.

(Observed values are total observed shift minus normal mass shift)

Level	$^3S_1$	$^1S_0$
Observed		
Fred *et al.**	0.141 K	0.099 K
Bradley & Kuhn (17)	0.10±0.02	0.10±0.02
Calculated, Stone (20)		
from Huang functions	0.076	0.159
from corrected Hylleraas-Undheim functions	.0758	0.149

* Dr. Fred, in a private communication, has kindly deduced level shifts from the line shifts reported (18).

considerably larger than that predicted by the Hughes-Eckart theory and a positive residual in the ^2S term series that may be attributable to polarization. No modern calculations are available on $_3$Li I.

In no spectrum of any heavier atom has the mass shift been studied for as long series as in neutral helium and lithium. The magnitude of each of the contributions varies approximately as the inverse square of the mass until for $Z\sim30$ it becomes difficult to detect. Several calculations have been made for heavier species, but all need recalculation with improved wave functions.

An important series of attempts has been made by Crawford and his students (27 to 30) to follow the exchange mass shift in increasingly heavy atoms. As illustrated above in helium, each exchange mass term has opposite signs for the triplet and the singlet in configurations with two electrons outside closed shells. This difference can be used to distinguish the specific mass effect from the field effect, which is, in any spectrum, to a close approximation a function of the electron configuration only. In a configuration $nsnp$ the average wave number difference between isotopes differing by ν units in N may be expressed with the aid of a parameter k:

$$\nu^{-1}[(\text{singlet minus triplet})_{N+\nu} - (\text{singlet minus triplet})_N] = \frac{2Rm_r}{\bar{A}^2 m_1} k(ns, np),$$

where R is the Rydberg constant, m_1 is one-sixteenth the mass of the O^{16} atom, $\bar{A}$ is the average atomic weight of the isotopes in a pair differing by ν units in N, and k is a function of the principal quantum number n. Crawford and his group have found k to be almost independent of n. With the aid of information about k it has been possible to subtract off approximately the specific as well as the normal mass effect from the observed total shift in some cases, among heavy atoms, in order to isolate the contribution of the field effect.

3. FIELD EFFECT

A system consisting of an electron and a point nucleus possesses a certain Coulomb energy. If the electron has an appreciable probability density at radii down to zero, as in the case of an s-electron, then the energy of the

system is higher if the nucleus is spread out, for instance over a sphere with radius R, than if it were concentrated at a point. This consideration is not essentially changed by the presence of other electrons disposed with approximate spherical symmetry about the nucleus, except for screening by other s-electrons. The structure of the nucleus was suggested by Goudsmit (31) as the basis for the isotope shift in the spectra of heavy atoms. The suggestion was implemented with qualitative success by Bartlett (32) through a (not altogether correct) calculation of the shift between isotopes with nuclei of the same density, i.e., in the case $R=R_1A^{1/3}$, where R_1 is a constant and A equals $Z+N$, the atomic weight integer for the isotope. Between two isotopes, the Coulomb cutoff energy is higher for the (heavier) one with the greater radius, and from first-order perturbation theory it may be seen that the lighter system has the lower energy, in agreement with observations on heavy nuclei of Figure 1c. This volume effect is recognized as the principal part of the field effect.

The magnitude of the calculated volume effect depends somewhat upon the radial charge distribution in the nuclear model chosen; the influence of the charge distribution has been studied by Humbach (33) with the aid of a parameter introduced by Rosenthal & Breit (34). For any assumed approximate radial dependence, in general, the value of R_1 determined from the experimental data depends upon what function of the radius is being measured. The volume effect in the isotope shift between atoms (Z,N) and $(Z, N+2)$ is a measure of $\langle r^{2\rho}\rangle\ (Z,\ N+2)-\langle r^{2\rho}\rangle\ (Z,\ N)$, where the brackets $\langle\rangle$ stand for an average over the charge, and $\rho=(1-\alpha^2Z^2)^{1/2}$, α being the fine-structure constant. The quantity $\langle r^{2\rho}\rangle$ also occurs in the Coulomb scattering of high-energy electrons. The current best value of R_1 pertinent to isotope shift studies is about 1.20×10^{-13} cm. (35, 36a); any direct evaluation requires a consideration of nuclear deformation, which is discussed below. An estimate made purely on the basis of a spherical nucleus in which each nucleon always occupies the same volume would lead to a value of less than 10^{-13} cm. for R_1, which is unreasonably low compared with values from other data gathered in different fields. Low (36) has considered the consequences of the reduction in R_1 from earlier estimates that were in the neighborhood of 1.5×10^{-13} cm. Several expressions different from $R=R_1A^{1/3}$ have been proposed for the dependence of R upon A, and for the charge density distribution within the nucleus. The general problem of the charge distribution within the nucleus, as observed in a wide variety of experiments, has been reviewed by Ford & Hill (37).

The electron-neutron interaction potential gives rise to a shift (38), small compared with the volume effect and increasing more slowly with increasing Z, and smaller than it was expected to be when it was first noted (39).

Breit (40) has pointed out the invalidity of the naive idea, from size considerations, that there should be an odd-even staggering of the opposite sign from that observed. Possible contributions to the field effect on the part of nuclear polarization (41) and the intrinsic moment of the electron (42) have been pointed out by Breit and his students.

In one of the most important recent theoretical papers on isotope shift

Wilets, Hill & Ford (43), show that the observed undulation of the shift curve among the even-even isotopes, and also the observed odd-even staggering, may be accounted for in a degree approaching the quantitative, by the detailed consideration of the nuclear asphericity earlier emphasized more qualitatively in the pioneering work of Brix & Kopfermann (44, 45) They show, moreover, that the ratio of about two-thirds usually found between the observed isotope shift in the field-shift part of the atomic table and the shift calculated from the (incompressible) volume effect alone, may be expected as a result of nuclear compressibility (46, 47). Figure 2 shows the observed ratio as a function of N.

Broch (48) avoids simple perturbation theory in calculating the volume effect arising from the distortion of the electron wave function by the distributed nuclear charges. Bodmer (35, 49), developing Broch's method and using calculational techniques developed for the study of the Coulomb scattering of high-energy electrons, shows that the isotope shift is less than that calculated from first order perturbation theory by a factor varying from 1 for low Z to zero for $Z=1/\alpha$, and having a value of about 0.8 for $Z=82$ and 0.6 for $Z=100$. He also relates the isotope shift to the isotopic difference in nuclear deformation, which in turn is correlated with intrinsic quadrupole

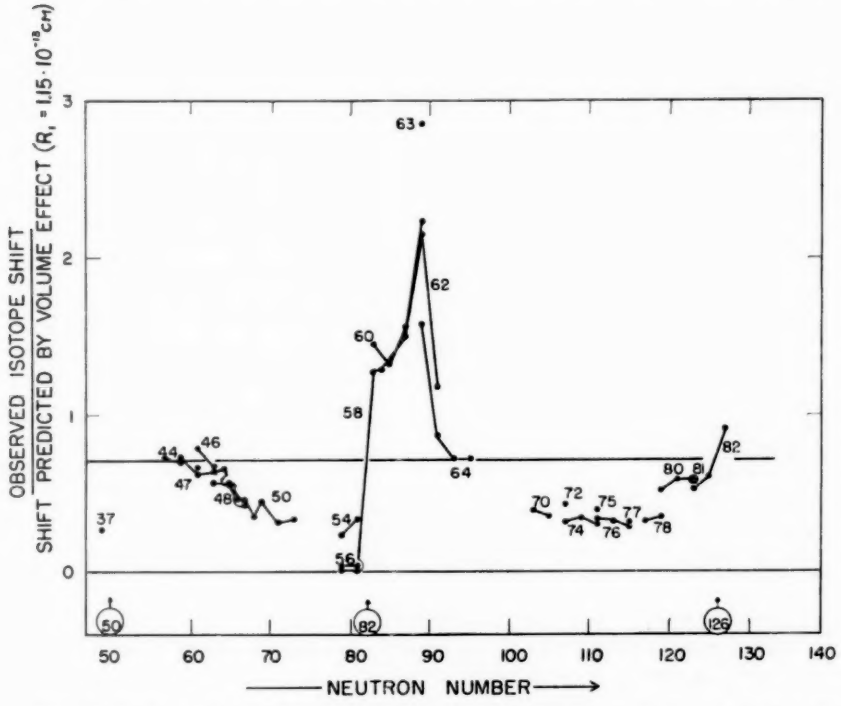

FIG. 2. Ratio of observed isotope shift to predicted volume effect as function of neutron number. A point at abscissa N refers to the shift between $N-1$ and $N+1$, except that the point at $N=84$ refers to the shift between $_{62}Sm^{144}$ and $_{62}Sm^{148}$.

moment, and shows independently of Wilets, Hill & Ford that differences in nuclear deformability lead to odd-even staggering with the correct sign. Ivanenko & Kolesnikov (50) emphasize the role of the closed shells in the isotope shift curve.

Electron screening effects have been studied in the isotope shift of spectrum lines, as in other phenomena, by comparing successive elements in the atomic table and successive stages of ionization in the same element (2, 51, 52).

Experimental work, and calculations or theoretical work (indicated by "Calc"), have been done on the isotope shift in the spectra of the following individual elements:

$_{1}$H (1)[2]		$_{48}$Cd (1, 29, 94 to 100)		
$_{2}$He (1, 17, 18)	Calc (19, 20, 21, 22, 25, 53)	$_{49}$In (101)		
		$_{50}$Sn (1, 30, 102, 103, 104)		
$_{3}$Li (1, 26)	Calc (6)	$_{51}$Sb (1)		
$_{5}$B (1, 54)	Calc (55)	$_{52}$Te (1, 75, 106)		
$_{6}$C (1, 56, 57, 60)	Calc (58, 59)	$_{54}$Xe (1, 107)		
$_{7}$N (1, 60)		$_{56}$Ba (1, 75, 108)	Calc (109)	
$_{8}$O (61, 62)	Calc (63)	$_{58}$Ce (1, 110, 111)	Calc (112)	
$_{10}$Ne (1, 64, 65)	Calc (66)	$_{60}$Nd (1, 113, 114, 115)		
$_{12}$Mg (1, 67, 68, 69, 70)	Calc (71)	$_{62}$Sm (1, 33, 116)		
$_{17}$Cl (1)		$_{63}$Eu (1, 117)		
$_{18}$A (1, 72)		$_{64}$Gd (118 to 122)		
$_{19}$K (1, 73)		$_{66}$Dy (123)		
$_{20}$Ca (74)		$_{68}$Er (124, 125)		
$_{22}$Ti (75)		$_{70}$Yb (1, 126, 127)		
$_{24}$Cr (75)		$_{72}$Hf (1, 128), 129, 129a)		
$_{28}$Ni (75)		$_{74}$W (1, 130 to 133)		
$_{29}$Cu (1, 76, 77)	Calc (78)	$_{76}$Os (1, 134, 135, 135a)		
$_{30}$Zn (1, 75, 77)	Calc (1, 27, 78, 79)	$_{77}$Ir (1, 135, 136)		
$_{31}$Ga (1, 79a)		$_{78}$Pt (1)		
$_{32}$Ge (75, 80a)		$_{80}$Hg (1, 137 to 143)	Calc (34)	
$_{34}$Se (1, 81)		$_{81}$Tl (1)	Calc (34, 144)	
$_{35}$Br (1, 82)		$_{82}$Pb (1, 145 to 148)	Calc (34, 149)	
$_{36}$Kr (1, 83)		$_{83}$Bi (150)		
$_{37}$Rb (1, 84)	Calc (78)	$_{84}$Po (151)		
$_{38}$Sr (1, 84a)		$_{90}$Th (152)		
$_{40}$Zr (1, 75, 85)		$_{92}$U (1, 153, 154)		
$_{42}$Mo (1, 75)		$_{94}$Pu (155, 156)		
$_{44}$Ru (86, 87)		$_{95}$Am (157)		
$_{46}$Pd (88, 89)		$_{96}$Cm (158).		
$_{47}$Ag (1, 90, 91)	Calc (92, 93)			

4. Acknowledgement

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[2] In this list, work that appears in Brix & Kopfermann's Landolt-Börnstein compilation (1) is cited only by reference to that compilation.

C. Y. Cho, J. G. Hirschberg, K. G. Kessler, H. Kuhn, K. W. Meissner, M. Robertson, K. M. Watson, and L. Wilets.

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PROPERTIES OF MEDIUM-WEIGHT NUCLEI

By K. Way, D. N. Kundu, C. L. McGinnis, and R. van Lieshout[1]
Nuclear Data Group, National Research Council, Washington, D. C.

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I. Introduction and General Conclusions

The perception and interpretation of systematic trends in nuclear properties have already resulted in considerable insight into nuclear structure It is with the hope of increasing the number of fruitful generalities that an-

[1] Present address: Institute for Nuclear Research, Amsterdam, Netherlands.

alyses of the currently known data on medium-weight nuclei have been made.

When the work was begun it was decided to designate as "medium-weight" those nuclei with mass numbers between $A=40$ and $A=140$. In this A-range, it was felt, nuclei would not show the highly individual features of the very light region where, as a rule, marked differences in most properties are found even among close neighbors; neither would they exhibit the clear signs of rotational behavior which in the heavy nuclei make for great similarity of properties over wide ranges of Z and N. The nuclei with A between 40 and 140 were expected to have similarity of properties over small stretches of Z and N with, of course, marked changes at the magic numbers in the ground-state spins, electric quadrupole moments, and binding energies. In fact, this is the region where it has been known for some time that magic number and shell effects stand out particularly clearly.

As the analyses progressed it became apparent that upper and lower cut-offs of $N=90$ and $N=22$, respectively, would have been better choices than $A=140$ and $A=40$ and that nuclei with N-values between these limits form a surprisingly coherent group. Such nuclei have the ground-state spins predicted by the single-particle shell model, and quadrupole moments not very much larger than this model would lead one to expect. In contrast to this most of the odd-A nuclei with $N>90$ have ground-state spins not predictable from the simple shell picture and quadrupole moments an order of magnitude larger.

However, it is in the properties of the first and second excited states of even-even nuclei that the most distinctive features are found. As already pointed out by Scharff-Goldhaber & Weneser (1) the ratio of the energies of these two states, E_2/E_1, is $\sim$2.2 in the medium-weight range while for heavy nuclei it is $\sim$3.3. For very light nuclei the ratio fluctuates widely.

Other even-even properties have been found to change with E_2/E_1. As is shown in Section V, the energy of the first excited state, E_1, when multiplied by A, has the fairly constant value of $\sim$60 Mev for the 2.2 -ratio nuclei, $\sim$15 Mev for the 3.3-ratio group, and $\sim$35 Mev for the group with the fluctuating ratio. The comparative lifetime, L, of this first excited state is also characteristic, being about 0.04 times the single-particle *proton* value, L_{sp}, for the 2.2-ratio groups; much smaller, $\sim$0.003 L_{sp}, when the ratio is 3.3; and much larger, $\sim$0.2 L_{sp}, for the light group with no characteristic ratio. A qualitative correlation between these lifetimes and the reciprocals of the static quadrupole moments can be noted.

When one comes to look for the exact boundaries of the medium-weight group, one finds that a dividing line from the light group at $N=22$ seems to have some real significance since for this and smaller N's the 2.2 ratio is never found. A close examination of the data also shows that a fairly sharp change at the upper boundary seems to come at neutron number $N=90$. For example for $_{62}Sm_{88}^{150}$, $_{64}Gd_{88}^{152}$, E_2/E_1 is 2.3 and 2.2 respectively while for $_{62}Sm_{90}^{152}$, $_{62}Gd_{90}^{154}$, it is 3.1 and 3.0. Recent work (1a) has shown that Eu^{152}

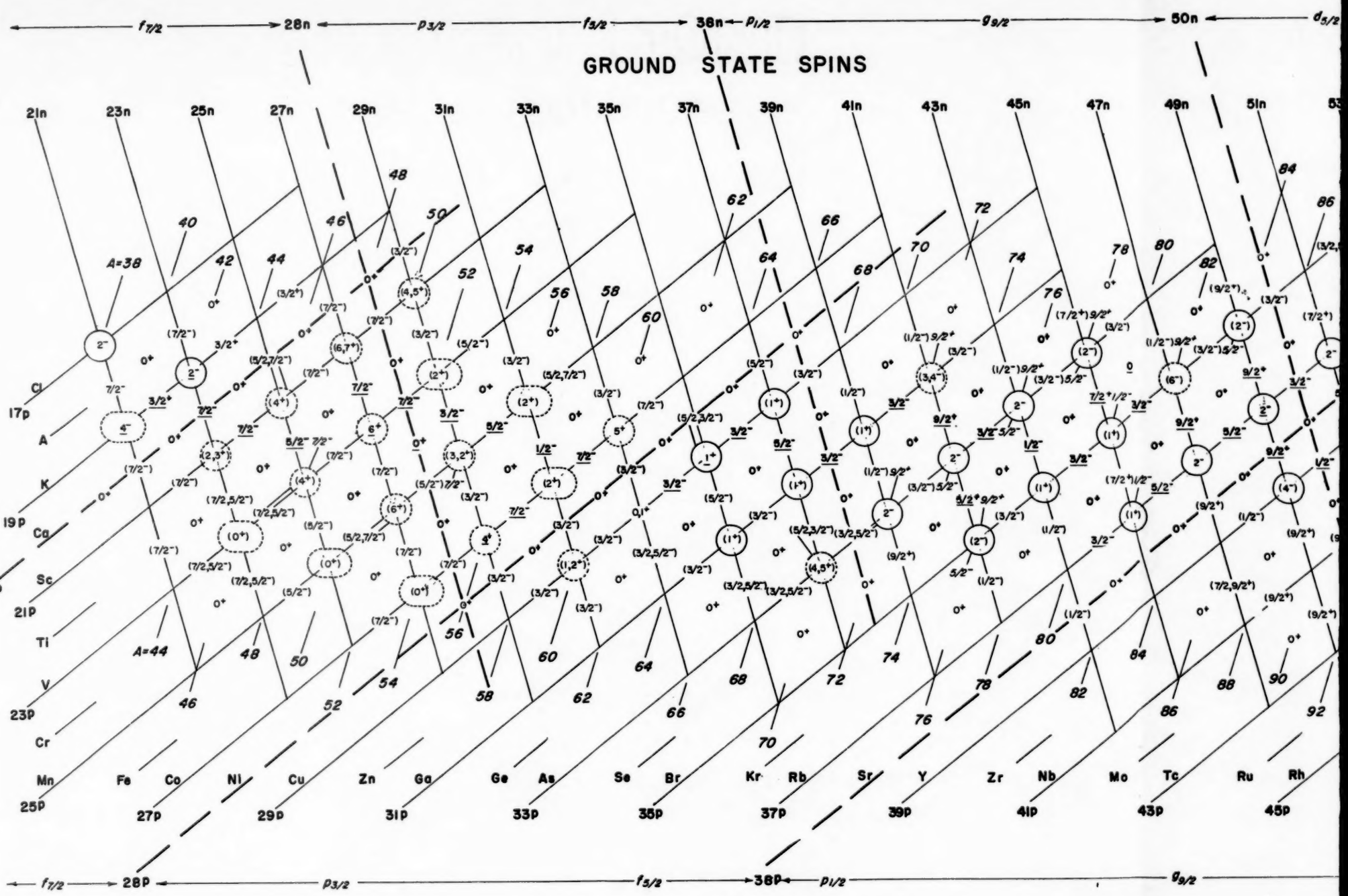
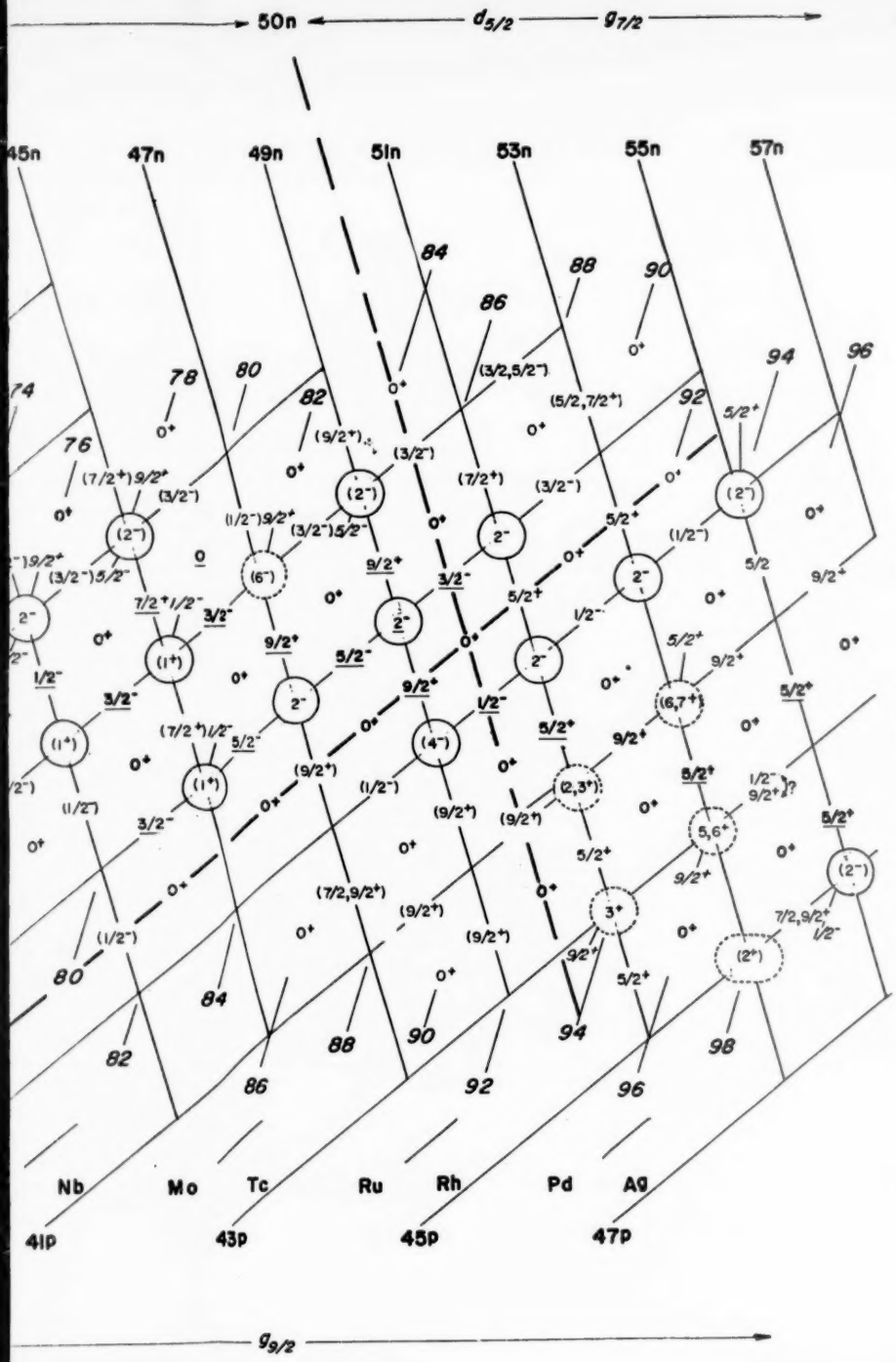
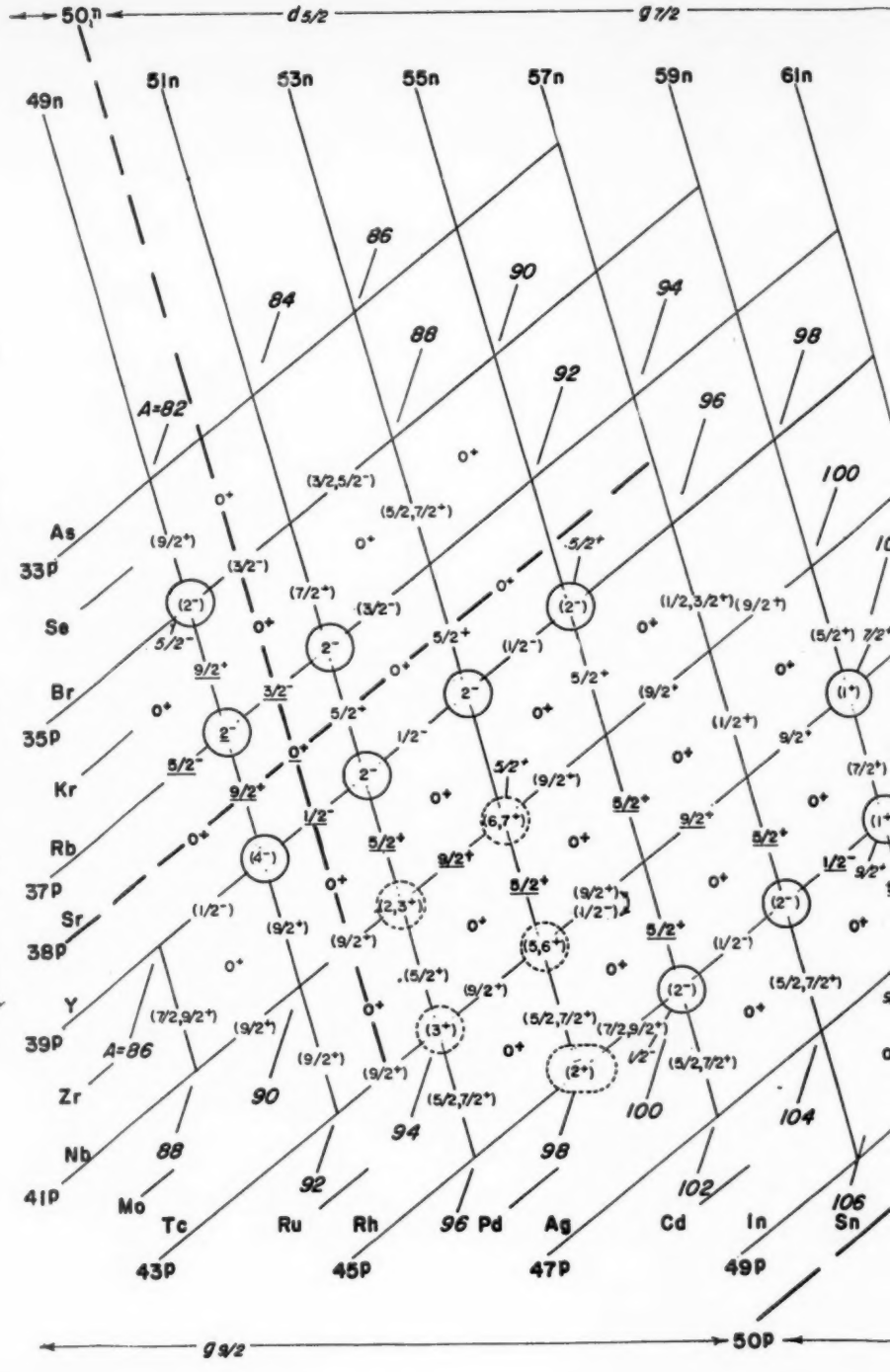

GROUND STATE SPINS
f7/2
28n
p3/2
f5/2
38n
p1/2
g9/2
50n
d5/2
21n
23n
25n
27n
29n
31n
33n
35n
37n
39n
41n
43n
45n
47n
49n
51n
17p
19p
20p
21p
23p
25p
27p
29p
31p
33p
35p
37p
39p
41p
43p
45p
Cl
A
K
Ca
Sc
Ti
V
Cr
Mn
Fe
Co
Ni
Cu
Zn
Ga
Ge
As
Se
Br
Kr
Rb
Sr
Y
Zr
Nb
Mo
Tc
Ru
Rh
A=38
A=44
f7/2
28p
p3/2
f5/2
38p
p1/2
g9/2

FIG. 1a


50n
$d_{5/2}$
$g_{7/2}$
45n
47n
49n
51n
53n
55n
57n
41p
Nb
43p
Mo
45p
Tc
47p
Ru
Rh
Pd
Ag
$g_{9/2}$


50n
$d_{5/2}$
$g_{7/2}$
49n
51n
53n
55n
57n
59n
61n
A=82
33p
As
35p
Se
37p
Br
38p
Kr
39p
Rb
41p
Sr
Y
A=86
Zr
Nb
Mo
Tc
Ru
Rh
Pd
Ag
Cd
In
Sn
$g_{9/2}$
50p

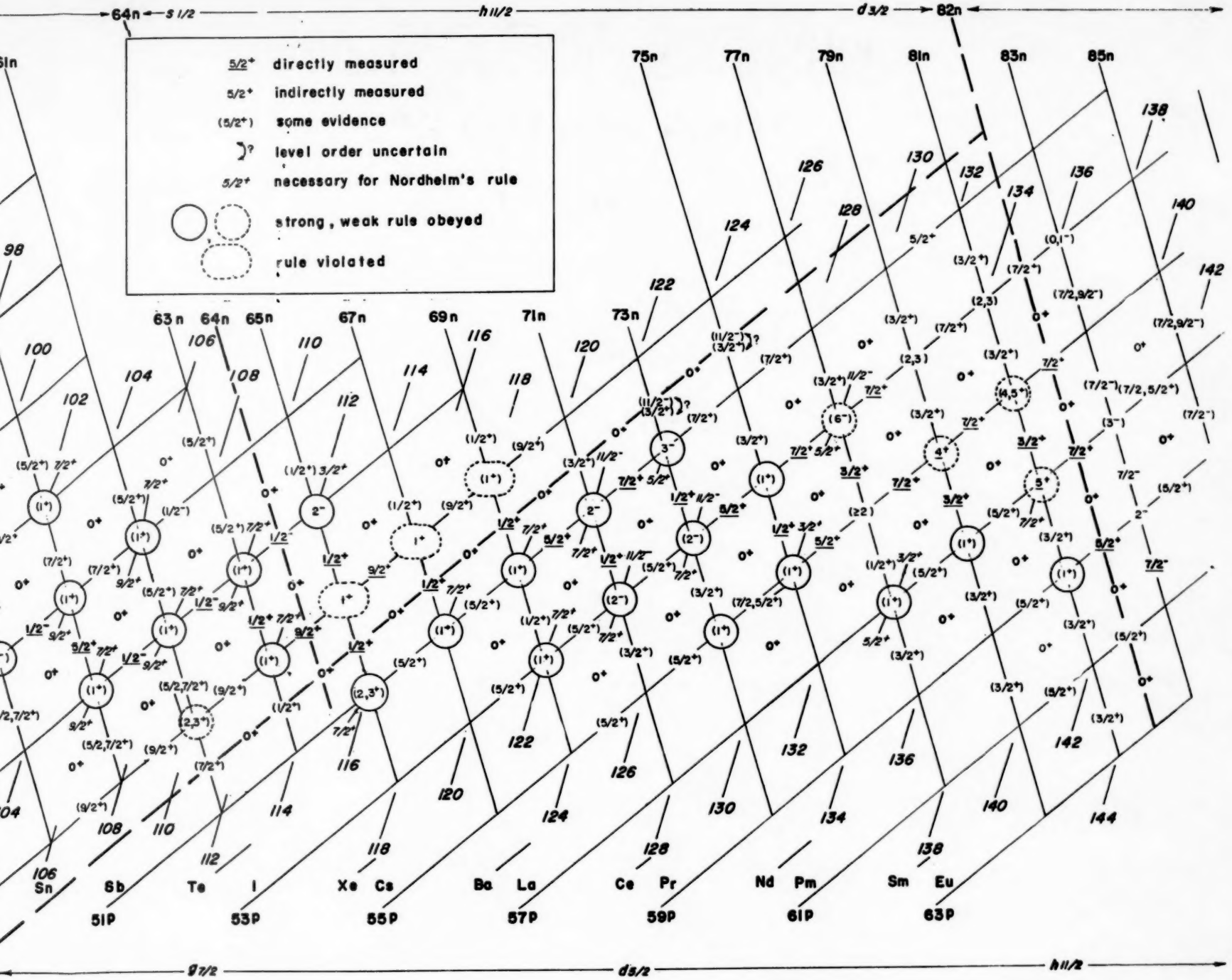

64n
s 1/2
h 11/2
d 3/2
82n
5/2+ directly measured
5/2+ indirectly measured
(5/2+) some evidence
level order uncertain
5/2+ necessary for Nordheim's rule
strong, weak rule obeyed
rule violated
63n 64n 65n 67n 69n 71n 73n 75n 77n 79n 81n 83n 85n
98 100 102 104 106 108 110 112 114 116 118 120 122 124 126 128 130 132 134 136 138 140 142
Sn Sb Te I Xe Cs Ba La Ce Pr Nd Pm Sm Eu
51p 53p 55p 57p 59p 61p 63p
g 7/2
ds/2
h 11/2

FIG. 1b

with 89 neutrons is more closely akin to the medium-weight than to the heavy group.

For some higher values of N, namely $N=114$ to 134 inclusive, a ratio of ~ 2.2 appears again. In this N-region the value $E_1A \simeq 75$ Mev is found and L for the first excited even-even state is $\sim 0.035\ L_{sp}$, a value very close to that for the lower N region. Shell effects also show up here for odd-A nuclei as is shown by the spins of the low-lying levels of Hg and Pb, the negative quadrupole moment of Bi^{209}, and the sharp changes in the binding energies after the magic number 82 for protons and 126 for neutrons. Thus we may say that this region exhibits medium-weight rather than heavy properties and speak about "medium-weight properties" which belong to two separated groups of nuclei, one with $22 < N < 90$, the other with $114 \leqq N \leqq 134$. These properties are listed and compared with those of other regions in Table I (see below).

This table perhaps does not bring out the fact that light nuclei can be characterized chiefly as a highly individual group. It is probably merely accidental that in it the ratio $E_2/E_1 = 2.2$ has not been found. The comparative lifetime, L, for the first excited even-even state is on the whole 5 times that for the medium-weight group, but great deviations about this value occur.

It seems curious that the light, medium-weight, and heavy groups show little or no difference in the amount of the deviations of the magnetic moments from the Schmidt lines as is pointed out in Section II. The data, however, are rather scanty for the light and heavy regions at the present time. Perhaps a better understanding of the spins in this region, as seems promised by studies of level order in deformed nuclei (2, 3), will put the heavy magnetic moment deviations in a new light. It will be interesting to see if the results for $M1$ lifetimes, as these become available, show the same independence of A.

TABLE I

CHARACTERISTIC PROPERTIES OF NUCLEAR GROUPS*

Property	Light $N \leqq 22$	Medium-Weight $22 < N < 90$, $114 \leqq N \leqq 134$	Heavy $90 \leqq N \leqq 112$, $136 \leqq N$
Odd-A Nuclei			
Ground-state J	J_{sp}	J_{sp}	not J_{sp}
Ground-state q	$\sim q_{sp}$	$\sim 3\ q_{sp}$	$\sim 30\ q_{sp}$
Even-Even Nuclei			
E_2/E_1	fluctuates	~ 2.2	~ 3.3
E_1A (no magic)	~ 35 Mev	~ 60 Mev	~ 15 Mev
$L(E2)$	$\sim 0.2\ L_{sp}$	$\sim 0.04\ L_{sp}$	$\sim 0.003\ L_{sp}$
Low-Level J's	0^+, 2^+, ?	0^+, 2^+, 2^+, or 4^+	0^+, 2^+, 4^+

* Subscript sp denotes values predicted by single-particle shell model, SPSM, see Appendix.

The distinguishing characteristics listed in Table I and some additional properties such as odd-odd spins, magnetic moments, odd-A low-lying levels, $L(E2)$ for odd-A nuclei, and $L(M1)$ are discussed in the following sections in the order shown in the outline. At the beginning of each section appears a list of the general statements that it now seems possible to make about the property in question, with special emphasis on its relation to medium-weight nuclei. Following this list is a graphical presentation of the data on which the general statements are based. Since most of the graphs are full-page, detailed explanations are given in the text in pargraphs headed, for example, "*Description of Figure 2, magnetic moments.*"

In general, references for the values plotted are to be found in *Nuclear Level Schemes* (4) or in the 1955 and 1956 cumulations "New Nuclear Data" in *Nuclear Science Abstracts* (5). In cases where different interpretations are possible, decisions are explained and references to the pertinent literature given in the "Description" paragraph.

Finally in each section there is an examination of the outstanding facts in the light of current nuclear models. In order to make this part of the discussion clearer, we give in an Appendix short descriptions of exactly what will be meant here by such terms as "single-particle shell model," SPSM.

II. Ground-State Spins

IIa. *General statements.*—The data on the ground-state spins for medium-weight nuclei show the strong shell features of this group. This fact, of course, has been from the beginning one of the strong supports for the shell model of Mayer & Jensen (6). These authors have summarized the spin results for stable nuclei and for radioactive nuclei with $A < 143$. In *Nuclear Level Schemes* (4) a new attempt was made to fit together all the known facts on β and γ intensities, log ft values, etc. The results of that work revised in the light of the latest data form the basis of the conclusions stated here. Classes of certainty for spin values were set up. These are described in Section IIb and indicated in Figure 1a and 1b, below.

The general statements that it is possible to make at present for nuclei with $21 \leqq N \leqq 81$ are the following:

1. For 223 stable and radioactive odd-A nuclei ground-state spins have been measured or can be plausibly assigned which, with 15 exceptions, agree with shell model assignments. These assignments are: for odd-nucleon numbers 21–27, $f_{7/2}$; 29–37, $p_{3/2}$ or $f_{5/2}$; 39–49, $p_{1/2}$ or $g_{9/2}$; 51–63, $d_{5/2}$ or $g_{7/2}$; 65–81, $s_{1/2}$, $h_{11/2}$ or $d_{3/2}$. The possibility of making firm dividing lines at 38 and 64 has perhaps not been emphasized before. Nine of the 15 exceptions occur for nuclei with 25 or 45 odd nucleons.

Table II. 1 shows the number of experimental values and the number of agreements for each of the different odd-nucleon regions and also gives the specific nuclei which have exceptional spin values. The data on which this table is based are given in Figure 1.

TABLE II.1

STATISTICS ON AGREEMENT OF EXPERIMENTAL ODD-A GROUND-STATE SPINS WITH SPSM VALUES

Particle Type and Number	SPSM Orbital	No. of Expt. Cases	No. of Agreements	Exceptions
p 21 to 27	$f_{7/2}$	19	15	${}_{23}V_{30}^{53}$ $(5/2^-)$, ${}_{25}Mn_{24}^{49}$ $(5/2^-)$, ${}_{25}Mn_{28}^{53}$ $(5/2^-)$, ${}_{25}Mn_{30}^{55}$ $(5/2^-)$
n 21 to 27	$f_{7/2}$	14	12	${}_{22}Ti_{25}^{47}$ $(5/2^-)$, ${}_{24}Cr_{25}^{49}$ $(5/2^-)$
p 29 to 37	$p_{3/2}, f_{5/2}$	27	27	
n 29 to 37	$p_{3/2}, f_{5/2}$	18	17	${}_{26}Fe_{31}^{57}$ $(1/2^-)$
p 39 to 49	$p_{1/2}, g_{9/2}$	30	29	${}_{45}Rh_{60}^{105}$ $(7/2^+)$
n 39 to 49	$p_{1/2}, g_{9/2}$	23	19	${}_{32}Ge_{45}^{77}$ $(7/2^+)$, ${}_{34}Se_{41}^{75}$ $(5/2^+)$ ${}_{34}Se_{45}^{79}$ $(7/2^+)$, ${}_{36}Kr_{45}^{81}$ $(7/2^+)$
p 51 to 63	$g_{7/2}, d_{5/2}$	31	31	
n 51 to 63	$g_{7/2}, d_{5/2}$	29	26	${}_{42}Mo_{57}^{99}$ $(1/2^+)$, ${}_{48}Cd_{63}^{111}$ $(1/2^+)$, ${}_{50}Sn_{63}^{113}$ $(1/2^+)$
n 65 to 81	$s_{1/2}, h_{11/2}, d_{3/2}$	32	32	

It is also possible to make a general statement about the odd-odd ground-state spins in the medium-weight region. Nordheim's rules (6a) state that if j_p and j_n are the resultant spins of the Z protons and N neutrons comprising an odd-odd nucleus (Z,N) respectively, then J, the ground-state spin, is given by:

$$\text{Strong rule: } J = |j_p - j_n|, \text{ if } j_p + j_n + l_p + l_n = \text{even}$$

$$\text{Weak rule: } |j_p - j_n| < J \leqq j_p + j_n, \text{ if } j_p + j_n + l_p + l_n = \text{odd}$$

It has been obvious for some time that, if j_p and j_n were assumed to be the ground-state spins of the odd-A nuclei $(Z-1,N)$ and $(Z,N-1)$ respectively, Nordheim's rules do not in general hold, even if j_p and j_n have expected shell model values.

The approach taken here is to consider a large group of nuclei composed of one of the proton and one of the neutron regions shown in column 1 of Table II.1, say the proton region 29 to 37 and the neutron region 39 to 49. The possible proton orbitals for this group are $p_{3/2}$ and $f_{5/2}$ and those for the neutrons are $p_{1/2}$ and $g_{9/2}$. Nordheim's rules then give $J=1^+$ for the combination $(p_{3/2})(p_{1/2})$; for $(f_{5/2})(g_{9/2})$, $J=2^-$; for $(p_{3/2})(g_{9/2})$, $J=6^-$, 5^- or 4^-; and for $(f_{5/2})(p_{1/2})$, $J=3^+$. The rules then allow 1^+, 2^-, 3^+, 4^-, 5^- or 6^- for the group as a whole. Without the rules, but with the assumption that $j_p=3/2^-$ or $5/2^-$ and $j_n=1/2^-$ or $9/2^+$, in addition to the above J's, values of 2^+, 3^- and 7^- are to be expected. Without the rule and with no restrictions on the coupling, all J's from 0^+ to 5^+ and from 0^- to 17^- are possible. (The largest

spin that has been observed is ≥ 7 for Lu^{176}.) Among the 14 available experimental values for the above group, the only spin values found are 1^+, 2^-, 6^-, and in one case 3^- or 4^-. Thus with one possible exception, Nordheim's rules can be said to hold for the group as a whole. This same statement can be made of the other proton-neutron groups for which data are available.

2. For odd-odd nuclei with odd-proton number and odd-neutron number in one of the regions mentioned in Table II.1, i.e., 21 to 27, 29 to 37, etc., Nordheim's rules permit only certain spin values. In a total of 81 cases, these permitted values are found 69 times. Three of the 12 exceptions are 0^+ values for $Z=N$ nuclei, three are 2^+ values in group $Z=21$ to 27, $N=29$ to 37, where the 2^+ assignment may be questioned, and three are 1^+ values in the group $Z=39$ to 49, $N=65$ to 81. The other three exceptions are isolated cases, the most outstanding of which is K^{40}. These observations are summarized in Table II.2.

TABLE II.2

ODD-ODD SPINS PREDICTED BY NORDHEIM'S RULES

Proton Numbers	Neutron Numbers	Predicted J's*		Exceptions Observed†
		Strong Rule	Weak Rule	
17 to 19	21 to 27	$\underline{\underline{2^-}}$		$(4^-)^1$
21 to 27	21 to 27		$\underline{\underline{7^+}}$, $\underline{6^+}$, 5^+, $\underline{4^+}$, $\underline{3^+}$, $\underline{2^+}$, 1^+	$(0^+)^3$
21 to 27	29 to 37	1^+	$\underline{\underline{5^+}}$, $\underline{4^+}$, $\underline{3^+}$	$(2^+)^2$
29 to 37	29 to 37	1^+	$\underline{\underline{5^+}}$, $\underline{4^+}$, 3^+, $\underline{2^+}$	
29 to 37	39 to 49	$\underline{1^+}$, $\underline{2^-}$	$\underline{3^+}$, $\underline{4^-}$, 5^-, $\underline{6^-}$	
29 to 37	51 to 63	$\underline{2^-}$, $\underline{0^-}$	$\underline{6^-}$ to 2^-	
39 to 49	39 to 49	$\underline{\underline{4^-}}$	9^+ to 1^+	
39 to 49	51 to 63	$\underline{2^-}$, $\underline{1^+}$	4^-, 7^+, $\underline{6^+}$, $\underline{5^+}$, 4^+, $\underline{3^+}$	$(2^+)^1$
39 to 49	65 to 81	5^+, 3^+, 0^-	$\underline{5^+}$, $\underline{10^-}$ to 3^-, 2^-	$(1^+)^3$
51 to 63	65 to 81	$\underline{1^+}$, $\underline{3^+}$, $\underline{2^-}$	$\underline{\underline{5^+}}$, $\underline{4^+}$, 3^+, 8^-, 7^-, $\underline{6^-}$, 5^-, 4^-	$(3^-)^1$

* Values which have been observed experimentally are underlined. A line extending under two values means that the experimental number could be either one of the two. If the data can be interpreted by either a predicted or a nonpredicted value, only the first has been noted. There are 5 cases in which this occurred.

† The superscript shows the number of times the indicated spin value has been found.

The data for statements 1 and 2 are exhibited in graphical style in Figures 1a and 1b.

IIb. *Description of Figure 1, ground-state spins.*—This figure shows in two sections the ground-state spins, in units of $h/2\pi$, of nuclei with neutron numbers from $N=21$ to $N=82$.

Spins which have been directly measured by spectroscopic, microwave, nuclear induction, or similar methods are underlined. Spins measured indirectly through determination of β-ray spectral shapes, angular correlation,

or internal conversion coefficients of γ-rays are given without underscore. The 0 spins of the even-even nuclei have been included in this class. Spins on which there is some evidence from any other consideration are put in parentheses. Spins of stable nuclei are represented in bold-face numerals. There are a few nuclei for which ambiguity in the order of the levels makes the ground-state spin uncertain. In such cases the two possible values are shown with suitable arrows to indicate the doubt. An example is Tc^{97} where $9/2^+$ and $1/2^-$ levels are very close to each other and it is not definitely established which level is the lower one.

The spins of the isotopes of odd-proton elements are given on the lines slanting upward towards the right. Those for even proton elements are shown at the appropriate intermediate positions but are not connected by lines. Nuclei with constant neutron number, isotones, are connected by lines slanting to the left. The arrangement makes the spins of odd-odd nuclei appear at the corners of diamonds. The location of the numbers dividing the regions of Table II.1, i.e., 20, 28, 38, 50, 64, and 82 are shown for both neutrons and protons by heavy dashed lines. Odd-odd spins which obey Nordheim's weak rule in the manner described above are circled with dashes on Figure 1. Those obeying the strong rule have full circles about them. Odd-odd spins which disobey Nordheim's rules are surrounded by dashes in an oblate shape. In case a given spin could be accounted for by either the weak or the strong rule, the strong rule was arbitrarily chosen. In those cases where it was necessary to assign a spin to one or both of the odd-nucleon constituents other than that found experimentally for $(Z-1,N)$ or $(Z,N-1)$ in order to get agreement with Nordheim's rules, the assigned spins are shown in italics beside the measured spins. For example the 2^- spin assignment for $_{53}I_{71}^{124}$ can be found by Nordheim's strong rule if the 53rd proton is assumed to be in a $g_{7/2}$ state and the 71st neutron in an $h_{11/2}$ state. However, the experimental spin of $_{53}I_{70}^{123}$ is $(5/2^+)$ and the directly measured spin of $_{52}Te_{71}^{123}$ is $\underline{1/2}^+$. The italicized numbers $7/2^+$ and $11/2^-$ are therefore shown next to $(\underline{5/2}^+)$ and $\underline{1/2}^+$ respectively.

IIc. *Discussion of odd-odd ground-state spins.*—Among the numerous theoretical studies (7 to 12) of odd-odd ground-state spins, the most extensive are those of de-Shalit (7), Kurath (8), and Schwartz (12). Kurath and de-Shalit have investigated the spins of the low-lying levels of the odd-odd nuclei with one particle or one hole in both the neutron and the proton shells. They use Majorana (M) and Wigner plus Bartlett (W+B) forces respectively. Table II.3 shows the results for some different values of d, the ratio of the force range to the nuclear "size", and α, the ratio of B to W+B. Some nuclei just to the light side of the medium-weight region have been included. It is seen that the Majorana force of zero range is the most successful, giving the experimental value in eight out of nine cases. This agreement with experiment is much more pronounced than it was at the time of publication of Kurath's paper because of new and changed experimental data. Nordheim's rules fail here in four of the nine cases. Three of these fail-

TABLE II.3

GROUND-STATE SPINS OF ONE-PARTICLE OR -HOLE ODD-ODD NUCLEI

Nucleus	Expt. J	Calculated J				
		Nordheim	Kurath (M)		de-Shalit (W+B) $d=0$	
			$d=0$*	$d=10$	$\alpha=0.2$†	$\alpha=0$
$_{17}Cl^{34}_{17}$	(0^+)	$3^+, 2^+, 1^+$	0^+	3^+	$(3^+, 1^+)$‡	0^+
$_{17}Cl^{36}_{19}$	(2^+)	$3^+, 2^+, 1^+$	2^+	3^+	$(3^+, 1^+)$‡	0^+
$_{17}Cl^{38}_{21}$	2^-	2^-	2^-	5^-	5^-	2^-
$_{19}K^{38}_{19}$	(0^+)	$3^+, 2^+, 1^+$	0^+	3^+	$(3^+, 1^+)$‡	0^+
$_{19}K^{40}_{21}$	$\underline{4}^-$	2^-	4^-	5^-	5^-	2^-
$_{21}Sc^{42}_{21}$	(0^+)	7^+ to 1^+	0^+	7^+	$1^+(7, 3, 5^+)$‖	0^+
$_{21}Sc^{48}_{27}$	$(6, 7^+)$	7^+ to 1^+	6^+	6^+§	$1^+(7^+, 3^+, 5^+)$‖	0^+
$_{21}Sc^{50}_{29}$	$(4, 5^+)$	$5^+, 4^+, 3^+$	2^+	5^+ for $d=\infty$	$5^+(3^+)$‖	2^+
$_{27}Co^{56}_{29}$	$(4, 5^+)$	$5^+, 4^+, 3^+$	4^+		$5^+(3^+)$‖	2^+
$_{29}Cu^{66}_{37}$	1^+	$1^+, 2^+, 3^+, 4^+, 5^+$			1^+	1^+
$_{37}Rb^{86}_{49}$	$\underline{2}^-$	$1^+, 2^-, 3^+, 4^-, 5^-, 6^-$			7^-	2^-
$_{37}Rb^{88}_{51}$	2^-	$0^+, 2^-, 3^-, 4^-, 5^-, 6^-$			5^-	2^-

* $d=a/b$ where a and b are defined by $V=V_0e^{-r^2/a^2}$ and $R_l=N_{lt}r^le^{-r^2/b^2}$ respectively.
† $\alpha=B/(W+B)$.
‡ Degenerate levels.
§ 6^+ and 7^+ levels are degenerate at $d=\infty$.
‖ Next higher levels.

ures are for $N=Z$ nuclei for which other failures have already been noted and the fourth is K^{40}.

Schwartz (12) extended the treatment of de-Shalit to nuclei with more than one equivalent particle outside of closed neutron and proton shells. He found that a value of α could usually be found which would give the correct ground-state spin but that this value was not unique. For nuclei with neutrons and protons in the $f_{7/2}$ shell Schwartz's method gives the correct ground-state spins in five cases out of eight for $\alpha \leqq 0.01$. However, this value of α is quite different from that found by Schwartz, $1/6 < \alpha < 1/4$, to give good results for the nuclei examined by him.

III. MAGNETIC MOMENTS

IIIa. *General statements.*—It was first pointed out by Schmidt in 1937 (13) that magnetic moments of odd-A nuclei, when plotted against the nuclear spin, J, fall between the two sets of values obtained by assuming

the entire moment to be due to a single particle with J equal to $l+1/2$ and $l-1/2$ respectively. It was also noted that the values for any given J were not scattered between the boundary numbers but tended to clump into two rather well defined groups. The lines connecting the single-particle values are now generally called the Schmidt lines.

Almost all magnetic moments measured since the time of the first Schmidt plot have been found to fall into this same pattern. The clumps, however, are broadening out. At present there are even four values for light nuclei which fall just outside the Schmidt lines. Now that there is a considerable amount of data, it is found that the deviations from the Schmidt values show striking features themselves the most prominent of which are listed below. The deviations discussed are all those which consist in a displacement of the moments toward the center of the Schmidt plot and so do not include the four outward deviations mentioned above.

1. The magnitudes of the deviations from the Schmidt lines do not seem to vary in any consistent way with A. For example, for the odd-proton nuclei with $J=5/2$ which are nearest the $l+1/2$ limit the deviation is smallest for Pr^{141} and increases for each of the following in order: Al^{27}, Cs^{131}, Mn^{55}, Eu^{151}, Sb^{121}, Ta^{181*}, Re^{187}, Re^{185} and I^{127}. Moreover this order is not that of the quadrupole moments of these nuclei whose values are: -0.05, 0.16, . . . , 0.5, 1.2, -0.5, . . . , 2.6, 2.8, and -0.8 respectively.
2. The rule first proposed by Schawlow & Townes (14) that the magnetic moments of semi-mirrors lie at equal fractional distances between the Schmidt lines is valid only in a rather general way. Semi-mirrors are pairs of nuclei for which the Z of one is equal to the N of the other (e.g. ${}_5B_6^{11}$, ${}_4Be_5^9$). In 16 cases out of 25 the moments of semi-mirror pairs lie within 0.05 units of each other on a scale in which the distance between the Schmidt lines for both odd-neutron nuclei and odd-proton nuclei is taken to be unity. In four cases the separation is greater than 0.10.
3. The deviations from the Schmidt lines are nearly zero for nuclei for which $J=1/2$ and $l=1$. The deviations tend to increase with J for $l-1/2$ nuclei and to decrease for $l+1/2$ nuclei.
4. The magnetic moment of a nucleus formed by the addition of two neutrons or two protons to a given nucleus is closer to the Schmidt limit than is the moment of the original one provided the two nuclei have the same spins. There are six exceptions in a total of 30 cases to this rule which was first noted by de-Shalit (15).

The data on which these generalizations are based are shown graphically in Figure 2.

IIIb. *Description of Figure 2, magnetic moments.*—This figure shows the relation of measured magnetic moments, μ, of odd-A nuclei to the distance between the Schmidt lines. For every J this distance has been taken equal to unity and the values of the moments plotted in terms of the fraction

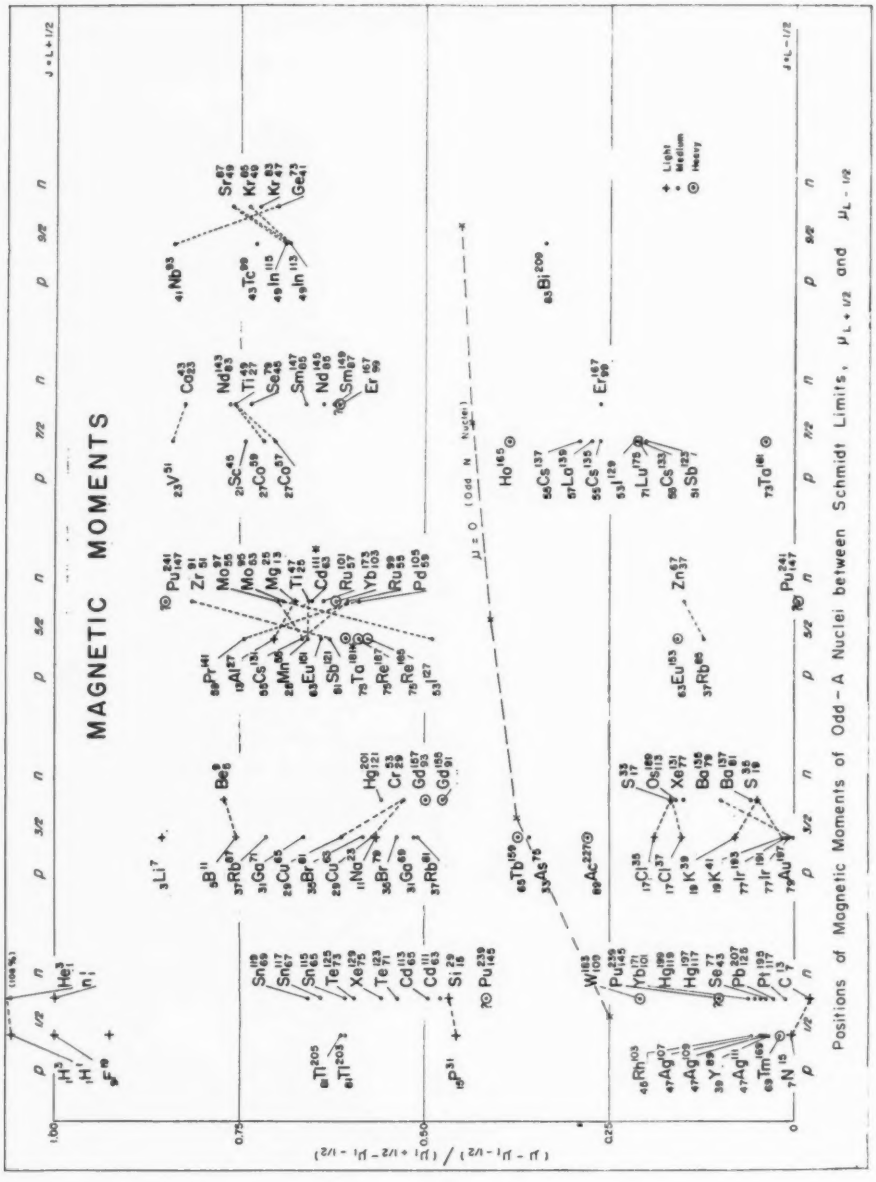

MAGNETIC MOMENTS
Positions of Magnetic Moments of Odd-A Nuclei between Schmidt Limits, $\mu_{L+1/2}$ and $\mu_{L-1/2}$
$\mu = 0$ (Odd N Nuclei)
+ Light
• Medium
◎ Heavy
$(\mu - \mu_{L-1/2}) / (\mu_{L+1/2} - \mu_{L-1/2})$
1.00
0.75
0.50
0.25
0
$J = L + 1/2$
$J = L - 1/2$
p 1/2
p 3/2
p 5/2
p 7/2
p 9/2
n

FIG. 2

$$(\mu - \mu_{l-1/2})/(\mu_{l+1/2} - \mu_{l-1/2})$$

where $\mu_{l-1/2}$ and $\mu_{l+1/2}$ are the Schmidt values for the J in question. This scheme makes it possible to show both neutron and proton moments on the same diagram and thus to compare them directly. The μ values of semi-mirrors are connected by dashed lines. Values for light, medium-weight, and heavy nuclei are distinguished by crosses, dots, and dots surrounded by circles respectively.

An asterisk after the symbol for the nucleus, e.g., Ta^{181*}, means that the value is for an excited state. Question marks indicate doubt about the sign of the moment. For Pu^{239}, Pu^{241}, and Er^{167} this was not determined by experiment (16, 17, and 18 respectively) so two values are given. Values for F^{19*}, Fe^{57}, Ni^{61}, and Np^{237} (19, 20, 21, 22, 23, respectively) were not plotted because of the size of the errors or doubt about the spins. The value used for Cd^{111*} ($\mu = 0.76$) was chosen as a weighted average between two values (24, 25).

IIIc. *Discussion of magnetic moments.*—The first observation points out that as far as magnetic moments go, the medium-weight nuclei are not in any way set apart as a group. The situation is certainly quite different from that for the electric quadrupole moments where the values for the heavy nuclei are an order of magnitude greater than they are for those of medium-weight. However, in spite of the fact that the magnetic moments do not help to differentiate the medium-weight group they will be discussed in some detail since a recent explanation by Arima & Horie (26) of the deviations from the Schmidt lines in terms of configuration mixing promises to throw great light on the actual way the neutron and proton shells fill up in the medium weight region.

A similarity in the moment deviations of semi-mirror pairs might be expected in view of the fact that the spins of the semi-mirrors are generally the same, a fact which suggests that the ground-state configurations of such nuclei are similar. It has been shown by de-Shalit (27) that the deviations from the Schmidt value of the magnetic moment due to an odd number of protons is

$$\Delta\mu^p = \mu \text{ (Schmidt)} - \mu^p = (g_l^p - g_s^p) f(j, J)$$

where $J = \Sigma_k j_k$. Similarly for the same number of odd neutrons

$$\Delta\mu^n = (g_l^n - g_s^n) f(j, J)$$

If now the even-type nucleons in semi-mirror pairs make no contribution to μ, then one would expect the ratio of the moment deviations to be

$$\begin{aligned} \Delta\mu^p/\Delta\mu^n &= (g_l^p - g_s^p)/g_l^n - g_s^n) \\ &= -4.59/3.82 = -1.20 \end{aligned}$$

The numerical value has been found by using the moments of the free protons and neutrons. Any "quenching" of these moments in nuclear matter will lead to a different result. Thus a constant ratio of -1.20 is to be expected

only if three conditions are fulfilled simultaneously, namely no quenching, identical coupling of the odd-type nucleons, and zero contribution of the even-type nucleons to the magnetic moment. From the way in which Figure 2 has been drawn it is easy to see, that if the -1.20 ratio held for semi-mirrors, the points for such pairs would have exactly equal ordinates on the graph. As already pointed out, this is not in general the case. The only conclusion possible is that the three conditions are not met simultaneously.

Ross (28) has argued that the effect on the magnetic moment due to modification of the intrinsic nucleon moments by the proximity of other nucleons should be roughly proportional to $A V_I/V_A$, where V_I is the interaction volume for a single nucleon and V_A is the nuclear volume. Since the latter is proportional to A, the effect is proportional to V_I, which should be approximately constant and presumably equal to the value it has for H^3 and He^3 where it amounts to about 0.2 nuclear magnetons. One notes, of course, that the effect in H^3 and He^3 acts like an enhancement and not a "quenching." If this effect is present in all nuclei it should produce deviations of just the opposite kind to the prevailing ones. Even if there is a marked change between light and heavy nuclei so that quenching actually does occur in the heavy region, it is, as Ross points out, hard to imagine that such an effect would produce moment deviations which vary greatly with small changes in nucleon numbers. However, a smoothly varying term may be present in addition to a fluctuating one as Russek (29) suggests.

If then it can be assumed that the major part of the deviations are due to causes other than quenching, the failure of the semi-mirrors to follow the Schawlow-Townes rule must be attributed to the failure of the odd-type nucleons to couple in the same way or of the even-type particles to contribute zero moment.

The possible contribution to the magnetic moment of a core which has part of the total angular momentum has been discussed by several authors (30 to 35). Obviously any sharing of angular momentum between the unfilled-shell particles and the core will lead, in general, for odd-proton nuclei to a decrease in the magnetic moment for both $J=l+1/2$ and $J=l-1/2$ since the orbital g-factor of the core instead of being unity, will be approximately Z/A. For odd-neutron nuclei the effect will be just the opposite since the core g-factor will be greater than the neutron orbital g-factor which is zero.

Bohr & Mottelson (30) calculate the correction to be expected from the strong-coupling collective model due to division of angular momentum between particle and core. For $J=l+1/2$ it amounts to one nuclear magneton (about 25 per cent of the separation of the Schmidt lines) and for $J=l-1/2$ to only about 0.1 n.m. (In this latter case for $J=3/2$ and $J=5/2$ the sign of the effect is opposite to that cited above for the general case.) The graph showing their results is given in the review paper of Blin-Stoyle (36). If the strong coupling picture were generally valid, greater moment deviations would be found for $l+1/2$ than for $l-1/2$. It is hard to see a clear-cut indi-

cation of this in Figure 2. Ford & Levinson (35), however, argue that such an effect is discernible for non-spherical nuclei.

The possibility that configuration mixing may have a decided effect on magnetic moments has been realized for some time. Preliminary discussions have been given, among others, by Davidson (37), Volkov (38), and Blin-Stoyle (39). Just recently a full treatment has been presented by Arima & Horie (26). By means of a perturbation calculation, using unfilled-shell particle interactions as the perturbing potential, these authors derive easily handled expressions for contributions to the magnetic moment due to configuration mixing for both the odd-type and even-type particles. Their results are given below for the three cases of importance. These cases and the meaning of the symbols used are illustrated in Figure 3.

For Case I the values of $d\mu$ due to configuration mixing are given by

$$d\mu = k_1 \frac{2j - k_2}{2j - 1} \frac{(l-1)l}{(2l+1)^2} (g_s - g_l)(-V_s I)/\langle \Delta E \rangle$$

For Case II

$$d\mu = -(k_1 - 1) \frac{(l+1)}{(2l+1)} \frac{(l+2)}{(2l+3)} (g_s - g_l)(-V_s I)/\langle \Delta E \rangle$$

For Case III, the values of $d\mu$ for the various possible sub-cases are given in Table III.1.

TABLE III.1

VALUES OF $d\mu$ FOR CASE III

Nucleus		$d\mu/[k_1(2j_2+1-k_2)/(2j_2+1)]$	Contributions from even numbers of
odd proton	$l+1/2$	$-\frac{(l+2)l_1}{(2l+3)(2l_1+1)} \times \begin{cases} (-V_s I)/\Delta E \times 4.585 \\ 1/2(V_s - V_t) I/\Delta E \times 3.826 \end{cases}$	protons neutrons
	$l-1/2$	$\frac{(l-1)l_1}{(2l+1)(2l_1+1)} \times \begin{cases} (-V_s I)/\Delta E \times 4.585 \\ 1/2(V_s - V_t) I/\Delta E \times 3.826 \end{cases}$	protons neutrons
odd neutron	$l+1/2$	$\frac{(l+2)l_1}{(2l+3)(2l_1+1)} \times \begin{cases} 1/2(V_s - V_t) I/\Delta E \times 4.585 \\ (-V_s I)\Delta E \times 3.826 \end{cases}$	protons neutrons
	$l-1/2$	$-\frac{(l-1)l_1}{(2l+1)(2l_1+1)} \times \begin{cases} (1/2(V_s - V_t) I/\Delta E \times 4.585 \\ (-V_s I)/\Delta E \times 3.826 \end{cases}$	protons neutrons

In these expressions V_s and V_t are the singlet and triplet interaction strengths of two nucleons. A δ-function potential is assumed. The values of $V_s I$, where I is the radial integral, are estimated from the experimental data on the pairing energy of equivalent nucleons in the manner of Mayer (39a)

and found to equal approximately $-25/A$ Mev. V_t is taken equal to 1.5 V_s. Values adopted for ΔE are based on $\Delta E=5$ Mev for the $d_{5/2}$, $d_{3/2}$ splitting in O^{17} and the assumption that splittings are proportional to $2l+1$ and $A^{-2/3}$. Approximately the same values for ΔE are used by Horie & Arima in their work on quadrupole moments (40) so that these values are tabulated in that paper also. There are not many experimental checks on the splittings at present. As shown in Section V on Levels, Nussbaum (41) finds fairly consistent evidence that $\Delta E(2p_{1/2}-2p_{3/2})\simeq 1.9$. This is to be compared with the Horie-Arima value of 1.5.

One notices immediately that the values of $d\mu$ given by the Arima-Horie expressions are always in the right direction to give the observed deviation

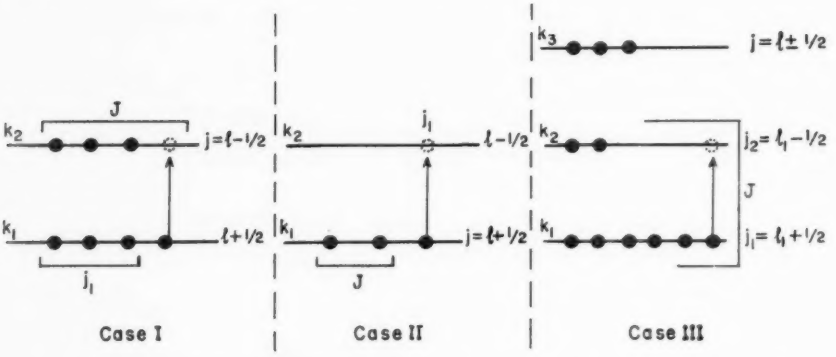


FIG. 3. Configurations which can contribute to magnetic moments. Ground-state configurations are shown by the filled-in circles. Excited-state configurations which when admixed with the ground state can make a significant contribution to the magnetic moment are indicated by arrows and dotted circles. k_1, k_2, k_3 are the numbers of particles in the indicated shells.

from the Schmidt lines. Moreover, their order of magnitude is easily found to be right. They account in a very natural way for the small deviations observed for $p_{1/2}$ nuclei since for such nuclei $d\mu$ vanishes (see Case I). Also it is obvious that, other things being equal, the $|d\mu|$ values for Cases I and II, which make the major contribution, increase with l for $l-1/2$ and decrease with l for $l+1/2$. Thus this theory accounts very well for observation (3) at the beginning of this section.

The theory can also account for small departures from the semi-mirror rule since it predicts different contributions to $d\mu$ from the configuration mixing of the even-type particles.

Arima & Horie have calculated values of μ to be expected from their theory for all nuclei for which experimental results are available. In general they used the ground-state configurations of Klinkenberg (42). The agreement with experiment is quite good. Only some typical results are given here in Table III.2.

TABLE III.2

VALUES OF MAGNETIC MOMENTS CALCULATED BY ARIMA & HORIE (26)

Nucleus	J	p Configuration	n Configuration	μ(calc.)	μ(exp.)
$_{29}Cu_{34}^{63}$	$3/2^-$	$(f_{7/2})^8 p_{3/2}$	$(p_{3/2})^4 (f_{5/2})^2$	2.17	2.23
$_{29}Cu_{36}^{65}$	$3/2^-$	$(f_{7/2})^8 p_{3/2}$	$(p_{3/2})^4 (f_{5/2})^4$	2.30	2.38
$_{24}Cr_{29}^{53}$	$3/2^-$	$(f_{7/2})^4$	$(f_{7/2})^8 p_{3/2}$	−0.49	−0.47
$_{33}As_{42}^{75}$	$3/2^-$	$(f_{5/2})^2 (p_{3/2})^3$	$(g_{9/2})^2$	2.21	1.44
		$(g_{9/2})^2 (p_{3/2})^3$	$(g_{9/2})^2$	1.62	
$_{51}Sb_{70}^{121}$	$5/2^+$	$(g_{9/2})^{10} d_{5/2}$	$(d_{5/2})^6 (h_{11/2})^6$	3.49	3.36
$_{40}Zr_{51}^{91}$	$5/2^+$	$(p_{1/2})^2$	$(g_{9/2})^{10} d_{5/2}$	−0.8	−1.3
$_{59}Pr_{82}^{141}$	$5/2^+$	$d_{5/2}$	$(h_{11/2})^{12}$	4.53	4.0
		$(g_{7/2})^6 (d_{5/2})^3$	$(h_{11/2})^{12}$	3.95	
$_{75}Re_{110}^{185}$	$5/2^+$	$(h_{11/2})^{12} (d_{5/2})^5$	$(i_{13/2})^4$	3.19	3.17
$_{75}Re_{112}^{187}$	$5/2^+$	$(h_{11/2})^{12} (d_{5/2})^5$	$(i_{13/2})^6$	3.17	3.20
$_{46}Pd_{59}^{105}$	$5/2^+$	$(g_{9/2})^6$	$(g_{7/2})^4 (d_{5/2})^5$	−0.45	−0.57
$_{21}Sc_{24}^{45}$	$7/2^-$	$f_{7/2}$	$(f_{7/2})^4$	5.62	4.75
		$(d_{3/2})^2 (f_{7/2})^3$	$(f_{7/2})^4$	4.74	
$_{71}Lu_{114}^{175}$	$7/2^+$	$(d_{5/2})^6 (h_{11/2})^8 (g_{7/2})^7$	$(f_{5/2})^4$	2.4	2.9
$_{49}In_{64}^{113}$	$9/2^+$	$(g_{9/2})^9$	$(d_{5/2})^6$	5.62	5.49
$_{49}In_{66}^{115}$	$9/2^+$	$(g_{9/2})^9$	$(h_{11/2})^2 (d_{5/2})^6$	5.59	5.50
$_{38}Sr_{49}^{87}$	$9/2^+$	$(p_{3/2})^4$	$(g_{9/2})^9$	−0.68	−1.09
$_{83}Bi_{126}^{209}$	$9/2^-$	$(h_{11/2})^{12} h_{9/2}$	$(i_{13/2})^{14}$	3.30	4.08

The success for Bi^{209}, though modest, is quite striking. The μ of As^{75} can also almost be accounted for if it is assumed that $g_{9/2}$ level is already filling here. The fact that $g_{9/2}$ levels ~0.4 Mev above ground are now known in both As^{73} and As^{75} makes such an assumption quite plausible. Agreement for Sc^{45} is also found if the proton configuration $(d_{3/2})^{-2}(f_{7/2})^3$ is assumed. Recent results for the level structure of this nucleus show that it is quite similar to that of Ca^{43} and not at all similar to that of Ca^{41}. King (43) has pointed out evidence in the low-lying levels of S^{37} and Cl^{39} for the break up of the 20 shell.

One sees thus that this elegant theory seems able to account for the moment deviations essentially on the many particle shell model, MPSM, and offers promise of distinct aid in ascertaining the order in which the shell-model levels are filled. Possibly it will need to be supplemented where the strong-coupling collective model seems particularly satisfactory.

One hurdle which the theory may or may not overcome is presented by the last of the four features noted at the beginning of this section, namely the rule that the addition of two protons or two neutrons to a nucleus pushes the

magnetic moment toward the Schmidt lines. The Arima-Horie picture gives such a result in general only where the two added particles go into an $l-1/2$ level. It is not always natural to assume that such is the case. However, the experimental change is small, <0.1 n.m. in 17 out of the 30 cases (see In and Re in Table III.2) while the change due to adding two nucleons in an $l-1/2$ level is generally several times this (see Cu in Table III.2). The fairly regular small changes may thus be explainable by some second order mechanism not considered in the theory at the present.

IV. Quadrupole Moments

IVa. *General statements.*—The ground-state spins of medium-weight nuclei were found to show shell effects very strongly. Shell effects are also clearly visible in the general Schmidt pattern of the magnetic moments and possibly also in the variations from this pattern if configuration mixing turns out to be the correct explanation for the deviations. That the quadrupole moments, q, also have an overall shell pattern was noticed by Schmidt in 1940 (44). Correlations of the data with the Mayer-Jensen shell model were made by Gordy (45), Hill (46), and Townes *et al.* (47) in 1949. Moreover, just as for the magnetic moments, it turns out that the q-values deviate considerably from the shell model predictions. The sign of the moment is often as expected but the absolute value is usually several times larger.

The single particle shell model, SPSM, gives $q=-\langle R^2\rangle(2j+1-2k)/2(j+1)$ where $\langle R\rangle^2$ is the expectation value of the square of the radial wave function for a single proton in the state j, and k is the number of particles in the shell of orbital j. According to this model the absolute value of the moment can never be greater than $\langle R^2\rangle(2j-1)/2(j+1)$. The sign of the moment should be negative when the shell is less than half filled and positive after the half-way point has been passed. No quadrupole moments are to be expected for odd-N nuclei.

The general statements that seem possible about medium-weight quadrupole moments at present are:

1. Negative moments appear for proton numbers 29, 41, 51, 53, and 59. (No value is available for Sc with $Z=21$.) A negative moment has been found for neutron number 41 for Ge^{73}. However, for Se^{75} which also has 41 neutrons a large positive moment is found. This is the nucleus for which an anomalous spin of 5/2 has already been noted.
2. The absolute values of the moments in the medium-weight region are in general less than a barn and usually only a few tenths of a barn. However, as noted in the Introduction there is good reason to count Eu^{151}, Ir^{191}, and Ir^{193} as "medium-weight" and for these nuclei q values of 1.2, 1.5, and 1.5 respectively are found. Thus a limit $q=1.5$ barns may be tentatively set for the medium-weight class. For heavy nuclei, q values of as much as 10 barns are found and for light nuclei the largest q is that of Al^{27}, namely 0.15 barns.

The quadrupole moments are shown graphically in Figure 4 where they

are also compared with SPSM values and with q's recently calculated (40) taking into account the possibility of configuration mixing.

IVb. *Description of Figure 4, quadrupole moments of light and medium-weight nuclei.*—The experimental values are shown in the upper part of the figure as a function of the number of odd nucleons. Values for nuclei with either odd numbers of protons or odd numbers of neutrons are included in the plot and are not distinguished from each other in any way. The dashed line has been drawn free-hand to show the general trend of the data. The indicated errors are experimental values as given by the authors reporting the measurements.

The crosses on the upper part of the figure are values calculated by Horie & Arima (40) in the manner described below.

At the lower part of the graph, q values predicted by the SPSM have been shown. These values were calculated assuming $\langle R^2 \rangle = 0.6 r_0^2 A^{2/3}$ where[2] $r_0 = 1.45 \times 10^{-13}$ cm. Average values of A were chosen for points at the beginning and end of each region.

For I^{127} the value of 0.75 of Murakawa (47a) was chosen as representing a reasonable average for the measurements of a number of different authors. A later value of 0.819 due to Jaccarino *et al.*, has been quoted by Livingston (47b) without error. The experimental value for Cd^{111*} (0.247 level, $J = 5/2$) is $\sim|1|$ (25). The positive sign was arbitrarily assigned.

IVc. *Discussion of quadrupole moments.*—If configuration mixing plays a role in magnetic moments it might be expected also to influence the deviations of the quadrupole moments from the shell-model values. Horie & Arima (40) have made a calculation of the deviations to be expected from configuration mixing in a manner entirely similar to that of their magnetic moment calculation. They find, of course, that the configurations which influence the q's are not in general the same as those affecting the μ's. Many more states now contribute, some giving a positive and some a negative correction to the shell-model value.

A study of Figure 4 shows that the quadrupole moments calculated in this way are not usually as large as the experimental values although in many cases considerable increases over the single-particle values are found. For instance, for Sb^{123}, Horie & Arima calculate $q = -0.39$, the SPSM value is -0.22, while the experimental value is -0.67 ± 0.10. The very large values found experimentally for the heavy nuclei are not given at all by such a calculation. This point has been emphasized by Perks (48). In this region an entirely different picture is obviously needed.

Configuration mixing gives quadrupole moments for odd-neutron nuclei. An interesting result is that for Ge^{73}. The experimental value is -0.2 ± 0.1. Horie & Arima calculate -0.43 for a ground-state neutron configuration of $(p_{1/2})^2 g_{9/2}$ and -0.27 for $(g_{9/2})^3$.

[2] This value for r_0 rather than the currently accepted one of 1.2×10^{-13} cm was used to facilitate comparison with the Horie-Arima calculations.

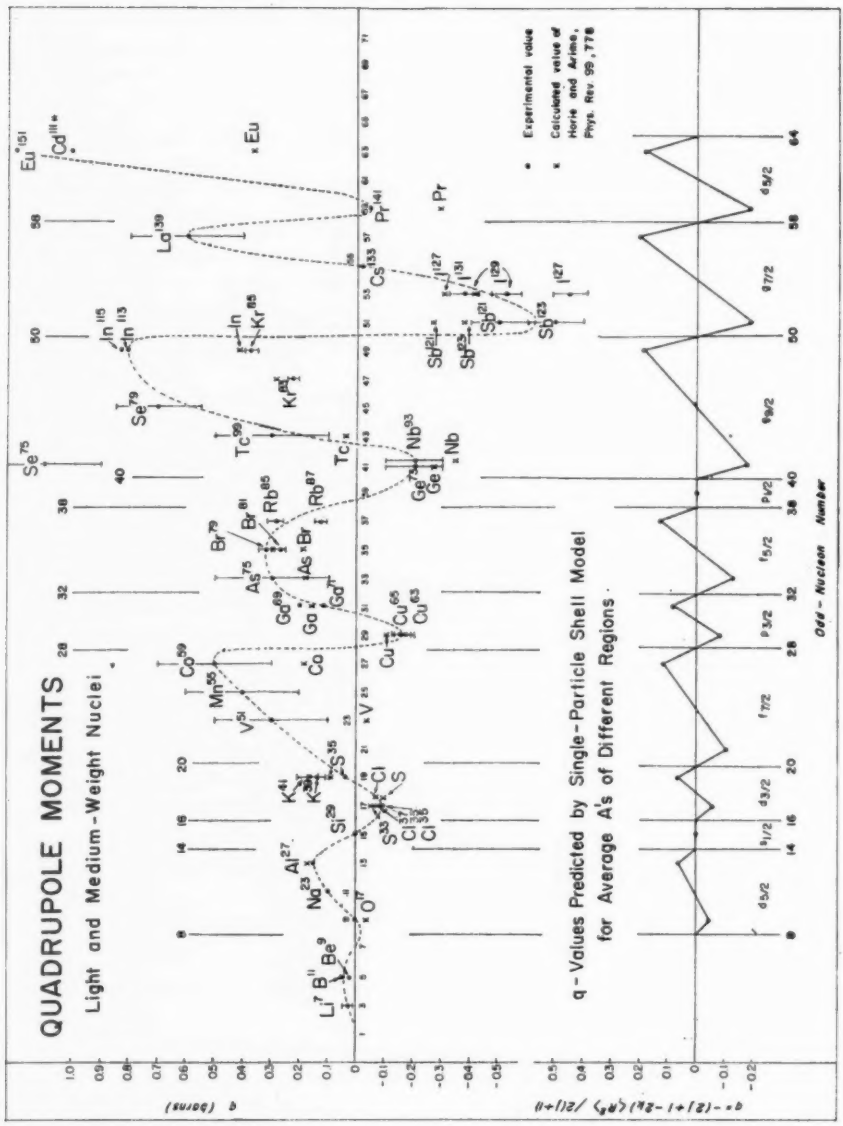

QUADRUPOLE MOMENTS
Light and Medium-Weight Nuclei
q - Values Predicted by Single-Particle Shell Model
for Average A's of Different Regions
Experimental value
Calculated value of Horie and Arima, Phys. Rev. 99, 778
Odd - Nucleon Number
q (barns)

FIG. 4

As the situation now stands, it seems possible that configuration mixing may account for most, if not all, of the magnetic moment deviations for nuclei of all weights. However, it can barely account for the quadrupole moment deviations of the medium-weight region and not at all for those of the heavy group.

V. Levels

Va. *General statements.*—The spins, magnetic moments, and electric quadrupole moments of medium-weight nuclei have been seen to show very marked signs of shell structure. Evidences of such structure also are visible in the patterns of the low-lying energy levels as is shown by the "islands of isomerism," in odd A nuclei which have been analyzed in an illuminating way by Goldhaber & Hill (49). Shell effects are also seen in the energies of the first excited even-even levels which, as Scharff-Goldhaber (50) first showed, have markedly high values at the magic numbers.

As already noted in the Introduction, a dramatic feature not obviously associated with the shell structure was recently pointed out by Scharff-Goldhaber & Weneser (1). Following their lead one can now state in the light of the presently available data that:

1. The ratio, E_2/E_1, of the energy of the second to the first excited state of even-even nuclei is $\sim$2.2 for nuclei with $22 < N < 90$ and $114 \leqq N \leqq 134$ while for $90 \leqq N \leqq 112$ and for $N \geqq 136$ this ratio is 3.3, the value to be expected for rotational states. It is mainly because of this fact that it has been possible to make clear a distinction between a medium-weight and a heavy group of nuclei.

A study of the distributions of E_1, the energy of the first excited state, for both even-even and odd-A nuclei has brought out additional differences between the medium-weight and heavy group as distinguished by the ratio criterion. The distribution of E_1A proved even more helpful. Distribution studies of this kind were previously made by Stähelin & Preiswerk (51). The results are:

2. For even-even nuclei with $22 < N < 90$ the energies of the first excited state fall into fairly distinct groups with peaks at 0.35, 0.6, 0.85, and 1.0 Mev. Nuclei which have either N or Z equal to 20, 28, 50, or 82 (20 cases) have first excited states at energies ranging from 1.1 to 1.9 Mev with no observable groupings. No levels occur below 0.3 Mev. In marked contrast to this, it is found that for nuclei with $N \geqq 90$ the first excited states energies form two groups, one with a peak at $\sim$0.1 Mev, the other with a broader peak at $\sim$0.4 Mev. In many cases the states of the first peak have been identified as rotational levels.
3. When the energies of the first excited states of even-even nuclei with $22 < N < 90$ are multiplied by A, the four groups mentioned in 2 coalesce into a single group with a peak at $E_1A = 60$ Mev. However, the values for nuclei with N or Z magic are spread out rather than

brought together by this treatment. The first class of level energies are thus approximately proportional to $1/A$ while the second are not. It should be noted that the power of A which brings about the coalescence is not uniquely determined. Nearly the same result is found if $A^{2/3}$ or $A^{4/3}$, instead of A, is used for the multiplication.

For the heavy nuclei with $N \geqq 90$ multiplication of the first excited state energy by A does not destroy the two peaks of the E_1 distribution. The one connected with the rotational level comes at $E_1A \sim 15$ Mev while the other is found at $E_1A \sim 75$ Mev which is just on the high side of big medium-weight peak at $E_1A = 60$ Mev. It was found that the nuclei whose values of E_1A fall in this second peak are just those in the Pb region, $114 \leqq N \leqq 134$, which show strong shell features, have moderate quadrupole moments and a value of E_2/E_1 of ~ 2.2. These nuclei thus have "medium-weight" rather than "heavy" properties.

4. For heavy nuclei, $90 \leqq N \leqq 112$, $N \geqq 136$, the spin of the second excited state is always 4^+, while for the medium weight group it is as often 2^+ as 4^+. The exact situation is shown in Table V.1. For nuclei with $24 \leqq N \leqq 88$ there are ten cases in which the spin of the second excited level could equally well be 2^+ or 1^+. If 2^+ is the correct assignment there are 18 2^+ levels in this group.

There is also a distinct difference between the distributions of E_1 and E_1A for odd-A nuclei with $N < 90$ and $N > 90$. However, at present no "diagnostic" features similar to the characteristic values of E_1A for even-evens are apparent.

5. For odd-A nuclei with $N < 90$ the spectrum of the energy of the first excited state shows two broad peaks, one at ~ 0.1 Mev and the other at ~ 0.6 Mev. The first may be illusory, caused by the fact that very low energy levels are missed through the experimental difficulties in observing them. The second is produced largely by nuclei for which the number of odd-type particles is magic plus or minus one. Nuclei for which the number of even-type particles is magic have widely different values and show no tendency to form a broad high-energy group as they do for the even-even nuclei. For $N > 90$, the odd-A E_1

TABLE V.1

DISTRIBUTION OF SPINS OF EVEN-EVEN 2ND EXCITED STATES

J	0^+	1^+	1^-	2^+	3^+	3^-	4^+	5^-	?	Total
$4 \leqq N \leqq 22$	3	1	1	3		2			7	17
$24 \leqq N \leqq 88$	2		1	8(18)		1	10		27(17)	49
$114 \leqq N \leqq 134$			4	6	1		4	1	2	18
$90 \leqq N \leqq 112$ / $136 \leqq N$							19			19

spectrum shows a strong low-energy line at ~0.05 Mev and a hint of one at ~0.4 Mev produced by nuclei in the Pb region.

6. When plots are made of the odd-A E_1A spectra, it is found that for $N<90$ the distribution falls off exponentially from the first peak at $E_1A=15$ Mev. If many low-lying levels have been missed the experimental curve may extend to $E_1A=0$. For $N>90$ there is a line as for the even-evens at $E_1A=15$ Mev. However, the trace of the Pb group has been diminished by the A multiplication rather than enhanced as it is in the even-even case.

Thus no order is apparent in the odd-A spectra of E_1 or of E_1A. However, regularities in the separation of levels of known character were pointed out some time ago by Hill (52) and Mitchell (53). As the data have increased, additional smooth curves, often parabolic, have been observed for the separations of levels whose spin and parity can be identified. In general these curves look very similar to those given in the 1952 review of Goldhaber & Hill (49) with one exception.

7. For odd-Z, even-N nuclei the parabolas for the $p_{1/2}$, $g_{9/2}$ separations now show extreme values at neutron numbers which increase as Z increases. Previously these appeared to come at $N=50$. For even-Z, odd-N nuclei the parabolas for $p_{1/2}$, $7/2^+$ separations show extreme values at $N=45$ as they did in the earlier plots. The data underlying all these observations are shown graphically in Figures 5, 6, and 7.

Vb. *Description of Figure 5, E (2nd excited state)/E (1st excited state) for even-even nuclei.*—The crosses indicate values for nuclei which have magic numbers for neutrons or protons or both, considering 20, 28, 50, 82, and 126 as magic. The circles show data for cases in which the spin of the second excited state is 0^+ rather than 2^+ or 4^+, the squares for cases in which this level has odd parity. Squares with crosses inside indicate both odd parity and one or two magic numbers.

Vc. *Description of Figure 6, energy distributions of first excited states.*—The full lines show the number of cases in which the first excited states of the indicated energy are found using the data for all nuclei except those with one or two magic numbers as defined in the last paragraph. The results for the magic number cases are dotted. All the histograms have been "smoothed" by the method described by Feenberg (54). Because of space problems the area under the E histograms was made a little smaller than the area under the corresponding EA histograms although the ordinate scales are the same.

Vd. *Description of Figure 7, level separations.*—In this figure two possible values for a single nucleus are shown by crosses connected by dotted lines. If it is assumed that the ground state of Tc^{97} is $p_{1/2}$, rather than $g_{9/2}$ as suggested by Cork *et al.* (55), and if the disintegration energy is 0.20 Mev as implied by the β systematics (56), then, for $\tau=10^4$ years, $\log f^1t=8.2$, which agrees well with values for other first forbidden unique transitions. This interpretation gives the point at the top of the parabola and also the upper point on the Tc dashed line.

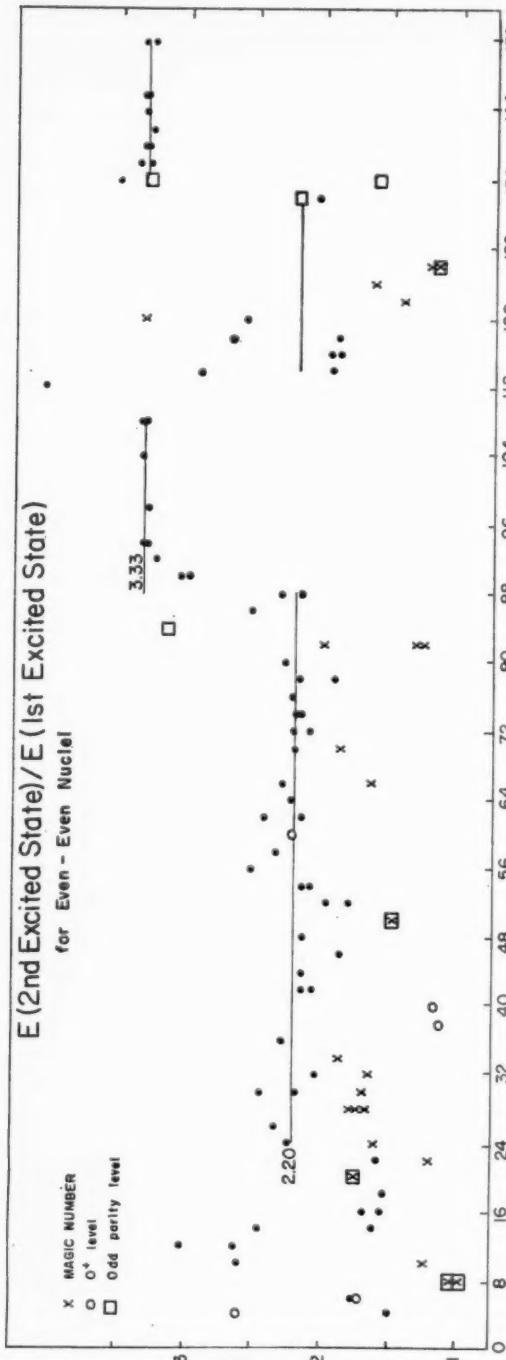

E (2nd Excited State)/E (1st Excited State)
for Even - Even Nuclei
MAGIC NUMBER
0^+ level
Odd parity level
3.33
2.20
$\frac{E_2}{E_1}$
NEUTRON NUMBER
0
8
16
24
32
40
48
56
64
72
80
88
96
104
112
120
128
136
144
152
1
2
3

FIG. 5

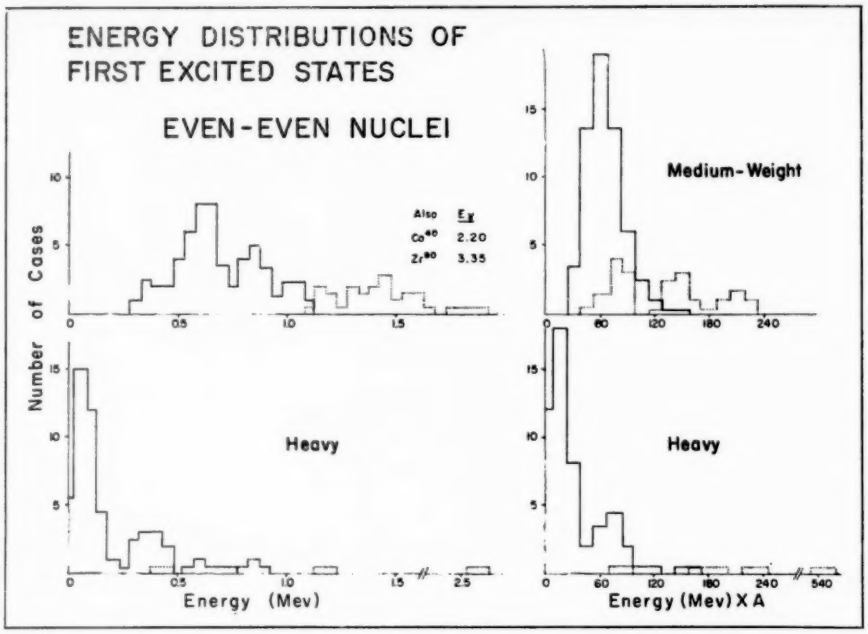


FIG. 6a. In this figure "Heavy" includes all nuclei with $N \geqq 90$.

Ve. *Discussion of even-even levels.*—Since the spin and the moment properties of medium-weight nuclei show strong shell features which in general can be accounted for by the single- or many-particle shell model, it is natural to look to this model for predictions of level spacing. For even-even nuclei there are no shell-model energy level calculations for the general case and therefore nothing at present which accounts for the ratio $E_2/E_1 = 2.2$ and the product $E_1A = 60$ Mev. Calculations do exist however, for the special case in which the neutrons or protons form a closed shell and the particles of the other kind do not. Here, results are in general agreement with experiment.

Stähelin & Preiswerk (51) suggested that the energy distribution of EA for the first excited state could be explained by configuration mixing. They pointed out that the mixing of the wave functions of many possible 2^+ states could result in a first excited state energy not very dependent on the number of particles in the unfilled shells. A closed shell for one type of nucleon would result in a marked decrease in the number of 2^+ states which would mix. Since there is an effective repulsion between the split 2^+ states, this would result in a rise of the energy of the lowest state. De-Shalit & Goldhaber (57) have given a very similar discussion. In addition, as Stähelin & Preiswerk remark, it can be argued on very general grounds that an $A^{-2/3}$ dependence would be expected.

Schwartz & de-Shalit (58) treated the interactions of like particles in a central field and found that to first order for a short-range attractive two-

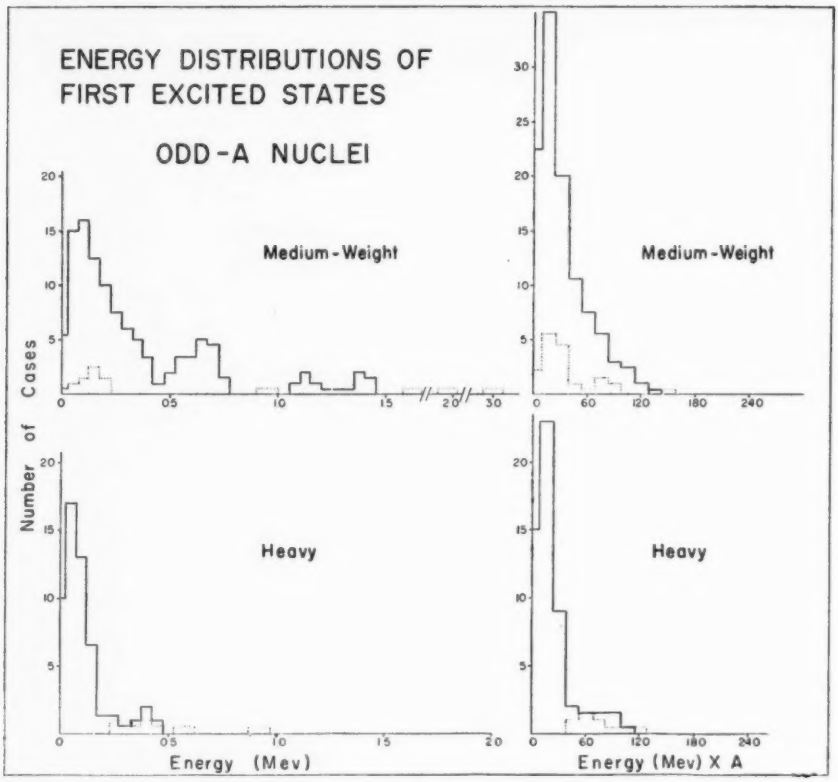


FIG. 6b. In this figure "Heavy" includes all nuclei with $N \geqq 90$.

body interaction with an arbitrary mixture of ordinary and spin-exchange forces, the energy difference between the ground state and the states of spin 2, 4, etc. with seniority 2 is independent of the number of particles in the unfilled shell. In addition they find that for two or four particles in the $f_{7/2}$ shell this same result holds for any two-body interaction whatever. These authors pointed out the fact that their results were not in agreement with the data for Ca^{42} and Ca^{44} whose 2^+ levels have energies of 1.53 and 1.16 Mev respectively. There are now available for comparison nine energy values for 2^+ states in the $f_{7/2}$ shell for the nuclei which have a closed shell for one type of particle. Only those for Ca^{44} and Ni^{62} differ significantly from the mean value of 1.43 for the other seven. Moreover possible energy assignments to 4^+ and 6^+ states show that the positions of these levels may also be independent of particle number. Table V.2 gives a summary of results for nuclei with one of the particle numbers equal to 20, 28, 38, or 50.

Kurath (59) has also found the Schwartz & de-Shalit (58) result for like particles in the $f_{7/2}$ shell. In addition he gives the separation of 2^+, 4^+, and

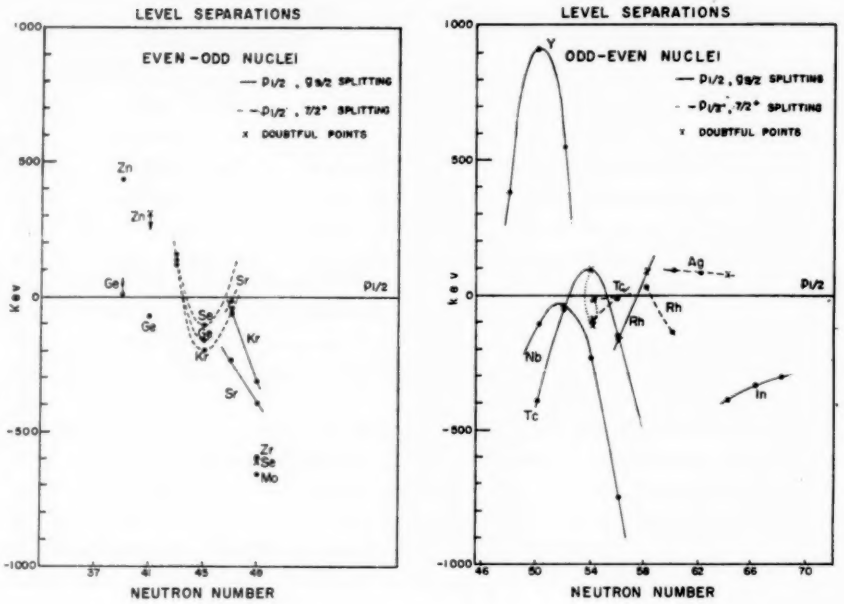


FIG. 7

TABLE V.2

ENERGIES OF 2^+, 4^+, 6^+ STATES IN SOME CLOSED-SHELL NUCLEI

Nucleus	Energy of Excited State*			Nucleus	Energy of First 2^+ State
	2^+	4^+	6^+		
$_{20}Ca_{22}^{42}$	1.45	(2.43)	(2.75 or 3.25)	$_{30}Zn_{38}^{68}$	1.02
$_{20}Ca_{24}^{44}$	1.16	(1.89 or 2.28)		$_{32}Ge_{38}^{70}$	1.04
$_{22}Ti_{28}^{50}$	1.58	(2.76)	(3.3)	$_{38}Sr_{48}^{86}$	1.08
$_{24}Cr_{28}^{52}$	1.45	2.43	3.13	$_{50}Sn_{64}^{114}$	1.30
$_{26}Fe_{28}^{54}$	1.41	(2.54 or 2.57)	(3.16 or 3.34)	$_{50}Sn_{66}^{116}$	1.27
$_{28}Ni_{30}^{58}$	1.45			$_{50}Sn_{70}^{120}$	1.18
$_{28}Ni_{32}^{60}$	1.33	2.51		$_{50}Sn_{72}^{122}$	1.14
$_{28}Ni_{34}^{62}$	1.17				
$_{28}Ni_{36}^{64}$	1.34				

* Parentheses indicate doubtful spin assignment.

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6^+ levels as a function of d, the ratio of the nuclear force range to the nuclear "size".[3] For $d=1.08$ one finds that $E(4^+)/E(2^+)=1.6$ and $E(6^+)/E(2^+)=2.3$, ratios which agree well with the data of Table V.2. However, this value of d is too big to give the experimental odd-odd spins of Table II.3. The crossover region between the two sets of values given there comes at about $d=0.4$.

Levinson & Ford (60) have estimated the contributions of configuration mixing to the energies of the 2^+, 4^+, 6^+ levels of Ca^{42} and find a considerable effect. However, they tried to get a fit to the assumption that the Ca^{42} level at 1.95 is the 4^+ level. If this level is instead a 0^+ state, as is indicated by other 0^+ data and if the 4^+ level is actually at 2.43 as is suggested in Table V.2, their conclusions would be somewhat modified.

Even if the even-even level patterns should be accounted for by the many-particle shell model, difficulties about the very fast transition rates would remain. These are discussed in Section VI.

Vf. *Discussion of odd-A levels.*— The fact that the first excited state of odd-A heavy nuclei lies at about the same position as that for even A (see Figure 6) can be accounted for by the rotational model. Aside from this there are no theoretical explanations of the experimentally found low-lying levels of odd-A nuclei. The existence of smooth curves for level separations, such as those of Figure 7 shows, however, that a certain amount of order does exist.

The success of the level separation analyses is attributable to the fact that the relevant states can be picked out selectively because of the occurrence of easily identified $E3$ and $M4$ transitions between them. Evidently one is not restricted to "single-particle states" in order to find regularities as is shown by the $p_{1/2}$,$7/2^+$ separation curve of Figure 7.

The deuteron stripping reaction is a powerful tool for the selection of levels of a particular character. The results can sometimes be supported or checked by means of capture γ intensities. A striking example of this is provided by the case of Ca^{41} for which it now seems possible to identify the level at 1.95 as $p_{3/2}$ and that at 3.9 as $p_{1/2}$ (61, 62).

Another good tool is to be found in γ-ray lifetime regularities such as those mentioned in the next section. For example the fast $E2$ transitions occurring in certain odd-A nuclei clearly identify levels which de-excite in a manner entirely unexpected from the single particle picture. Levels of this type deserve special attention in order to discover if they arise from the addition of a particle to the even-even core without destruction of the core's intrinsic structure. If so, several closely related states corresponding to different relative orientations of the last particle to the core may be expected.

Nussbaum (41) has made a special effort to identify the single-particle levels in the region $A=40$ to $A=60$. He finds that the present data do not indicate unique values for the level separations. There are, however, the following fairly consistent results for the indicated separations: $p_{3/2}$–$f_{7/2}$

[3] See Table II.3 for exact definition.

$\approx$1.6 Mev; $p_{1/2}$–$p_{3/2}\sim$1.9 Mev. For the separation $f_{5/2}$–$p_{3/2}$ there are two groups of possible values, one $\sim$1 Mev and the other $\sim$0 Mev. From these one gets estimates for the separation $f_{5/2}$–$f_{7/2}$ of 2.6 and 1.6. It is to be noted, however, that there is no experimental evidence from d,p angular distribution measurements for a single-particle $f_{5/2}$ level. In Sc^{49} the β-decay data show levels at 3.1 and 4.1. If it is assumed from the systematics of the region that $p_{3/2}$ is lower than $f_{5/2}$, a lower limit of 4.1 is given for the separation $f_{5/2}$–$f_{7/2}$ in Sc^{49}.

VI. Comparative Gamma-Ray Lifetimes

VIa. *General statements.*—Since most of the γ-ray lifetime measurements have been made for the medium-weight group, a comparison with values in the other regions is not possible at present. Only in the case of $E2$ transitions do comparable data exist for both medium-weight and heavy nuclei. In the statements below "medium-weight" will be taken to include the Pb-region nuclei mentioned in the Introduction.

Following Goldhaber & Sunyar (63) we define L, the comparative γ-ray lifetime, for electric transitions to be $\tau A^{2l/3} E^{2l+1}$ and for magnetic transitions to be $\tau A^{(2l-2)/3} E^{2l+1}$. Here τ is the mean γ-ray lifetime in seconds, A the mass number, l the multipole order of the transition, and E the γ-ray energy in Mev. Measured L values are compared with L_{sp}, the theoretical value for a single proton derived by Moszkowski (64). We have used $1.2A^{1/3} \times 10^{-13}$ cm for the nuclear radius and a statistical weight factor of unity in Moszkowski's formula. For a single neutron the values are about $(Z/A^{\frac{1}{3}})^2$ larger for electric and about 1.5 times larger for magnetic transitions (64). In terms of these quantities, the following general statements can be made:

1. For even-even $E2$ transitions, $L(\text{medium-weight})/L_{sp} \cong 0.04$ while $L(\text{heavy})/L_{sp} \cong 0.003$. For 80 per cent of the medium-weight cases, the deviation from the value given is not more than a factor of 2.3. For odd-A $E2$ transitions $L(\text{medium-weight})/L_{sp} = 0.08$. For 80 per cent of the cases the deviation from this value is less than a factor of five. Values of L near closed shells are larger than elsewhere for both even-even and odd-A nuclei.
2. In even-even medium-weight nuclei the second 2^+ state has anomalous decay properties in the light of the spin and parity selection rules. In three cases, Se^{76}, Sn^{116}, and Te^{122}, the ratio $E2/M1$ for the stopover transition has been measured and found to be about nine. From empirical transition probabilities one would expect $E2/M1$ $\sim$0.6.

   In 12 cases there is a measure of the relative intensity of the stopover to the crossover transition. If we assume the stopover is predominantly $E2$, then the ratio of the transition matrix elements, stopover to crossover, instead of being unity, varies from 8 to 250, averaging about 70.

Graham & Bell (65) have called attention to the fact that the $M1$

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transitions which the shell model designates as l-forbidden form two groups with specific L values. There are at present eleven such transitions known. However, for the $M1$ class as a whole (25 cases) the L values show a large spread.

3. For medium-weight l-forbidden $M1$ transitions, L(odd-neutron) $/L_{sp}=60$ and L(odd-proton)$/L_{sp}=300$.
4. For all $M1$ transitions, L(medium-weight)$/L_{sp}\cong 100$. Eighty per cent of the 25 cases differ from this value by less than a factor of 16.

A simple relation also seems to exist among a limited group of $E3$ transitions. As pointed out by Goldhaber & Sunyar (63), log L values for transitions between $p_{1/2}$ and $7/2^+$ levels when plotted against neutron number fall on two straight lines which slope away from $N=50$. However, a recent value for Ge^{77} is off their lines. The surprising thing is that there is very little difference between the comparative lifetimes for the odd-neutron and odd-proton nuclei in this group. For the $E3$ transitions as a whole $L/L_{sp}\cong 500$. The $E3$ transitions are in marked contrast to the M4 ones, which form the most uniform transition group yet known.

5. For $M4$ transitions $L/L_{sp}=7$. Eighty per cent of the 47 cases differ from this value by a factor of less than two. Moszkowski (66) has noted a tendency for the L's to decrease as a closed shell is approached.

VIb. *Description of Figure 8, comparative lifetimes for E2 transitions.*—Values of L as defined above are plotted for neutron numbers between 24 and 98 for transitions identified as $E2$. The horizontal line gives the value of L_{sp} as defined above.

Transitions in even-even isotopes of the same element are joined by straight lines. Odd-A points joined by a vertical line represent extreme values for different possible values of the excited state spin. Other lines between odd-A points join levels of the same spin and parity in the two isotopes of the element. The Coulomb excitation data is taken from the work of Temmer & Heydenberg (67), Stelson & McGowan (68), and Fagg *et al.* (69).

VIc. *Discussion of gamma-ray lifetimes.*—Goldhaber & Sunyar (63) presented in 1955 a very convenient classification of isomeric transitions in terms of comparative lifetimes, here called L. The accumulation of additional data since the closing date for their paper has not changed the main features of their analysis. In general measured L values are greater than the single proton estimate for all multipoles except $E2$.

The recent data show that while the small L, or fast $E2$'s occur throughout the entire periodic table, they are about ten times faster for the heavy than for the medium-weight group. Since the strong-coupling collective model has supplied an explanation for these transitions in the heavy region, it is natural to see if adaptations of this model can account for the medium-weight case. Two such adaptations have been proposed.

Wilets & Jean (70) suggest that strong coupling between the core and the unfilled-shell particles exists also in the medium-weight nuclei region but

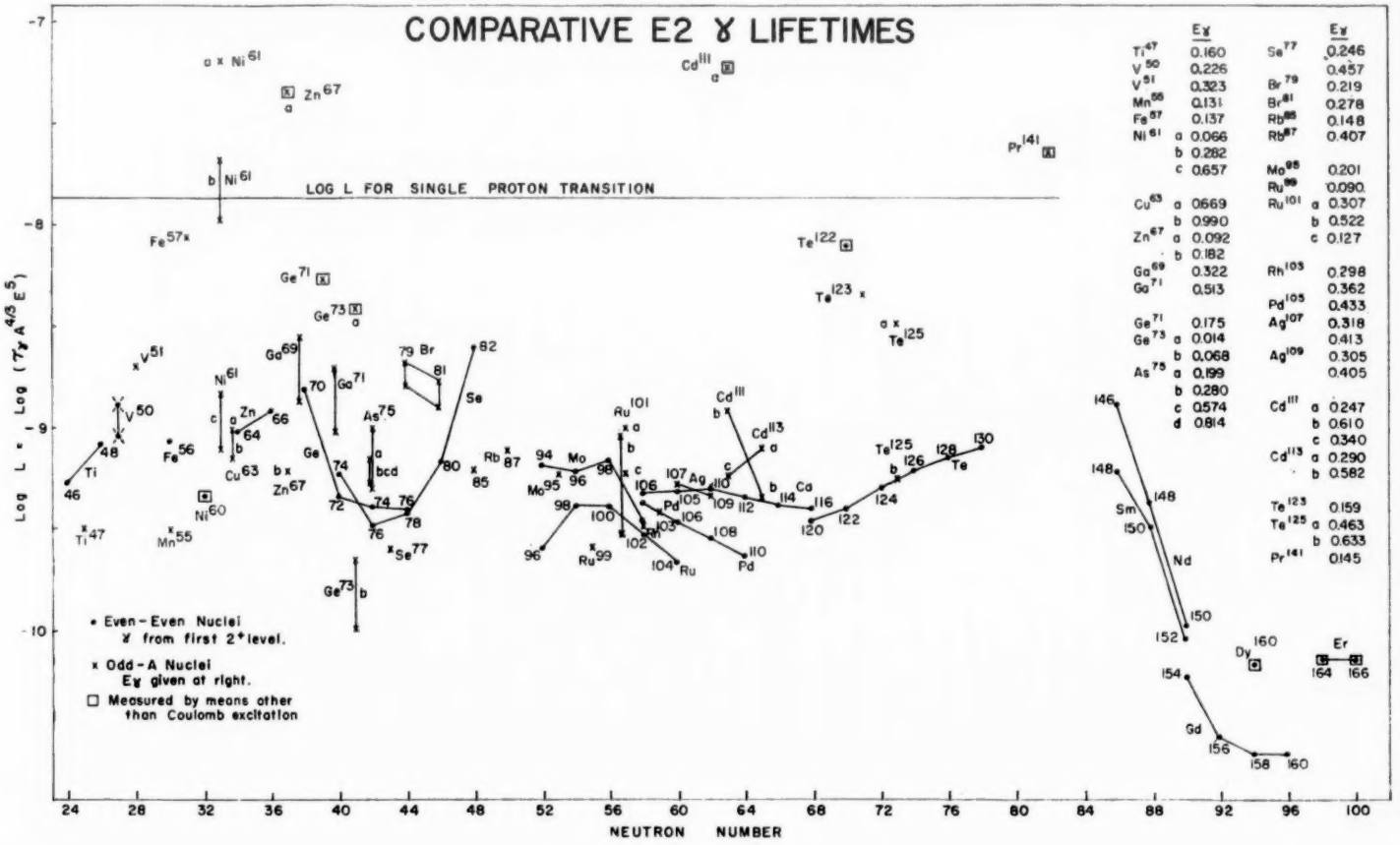

FIG. 8

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that such nuclei lack an axis of cylindrical symmetry, i.e., they are γ-unstable in terms of the definitions of the Appendix. With γ-instability large deformations are possible. The nucleus, however, will alternate between the oblate and prolate shapes and thus the spectroscopic quadrupole moment will be zero. The lowest modes of excitation will be the surface oscillations with energies $\hbar\omega$, $2\hbar\omega$, etc. The excitation phonons are bosons of spin 2 and the angular momenta of the first few levels are 0^+; 2^+; 0^+, 2^+, 4^+.

The collective feature of this picture accounts for the small L or the fastness of the $E2$ transitions. The ratio, E_2/E_1 is given as 2 in the first approximation. Moreover one expects the 2^+ to 2^+ stopover transition to be enhanced because of the favored nature of a one phonon jump. A ratio, $E_2/E_1 > 2$ and a raising of the 0^+ excited level above the 2^+, 4^+ pair is found from a more detailed treatment.

Scharff-Goldhaber & Weneser (1) propose weak to moderate rather than strong coupling. Their picture gives the same general results as that of Wilets & Jean. Both models predict a 2^+, 4^+ doublet at about twice the energy of the first excited level. Such doublets are known in a few cases, e.g. Ni^{60}, Kr^{82}, and possibly Mo^{96}, Cd^{110}, Cd^{114}, Sn^{116}, and Te^{124}. In the Scharff-Goldhaber & Weneser model the excited 0^+ level lies close to the 2^+, 4^+ state. Experimentally $_{46}Pd_{60}^{106}$ is the only nucleus known with a 0^+ level at $2.2E_1$ but here the second 2^+ level is at $3.0E_1$.

Attempts to find evidence in favor of one model or the other by way of electric monopole transitions (71) have so far not met with success.

In the single-particle model the matrix element vanishes for M transitions which are l-forbidden, i.e. which have an orbital angular momentum change greater than the multipole order. Following Jensen & Mayer (72), de Waard & Gerholm (73) have calculated extra-ordinary terms due to spin-orbit coupling and shown that a contribution proportional to $\Delta E/(2l+1)$ exists where ΔE is the spin-orbit splitting energy. Using recent measurements for the $d_{3/2}$–$d_{5/2}$ splitting in Cs^{133} and Cs^{135} they find that this spin-orbit contribution accounts for about 60 per cent of the $M1$ l-forbidden transition rate.

Sano (74) has been able to account partially for the fact that the $E3$ transitions in the odd-neutron nuclei have comparative lifetimes about equal to those of odd-proton nuclei rather than a factor of 10^8 larger as would be expected on the single-particle model. Using the weak-coupling model, and taking configuration mixing into account by means of simple perturbation theory, he has found $L_{calc}(\text{odd-neutrons})/L_{sp}$ of the order of 100 which is near the experimentally observed value. Actually his ratios of calculated to experimental lifetimes vary from about 2 to 60.

When the weak coupling calculation of Sano (74) is applied to the $h_{11/2}$–$d_{3/2}$ $M4$ comparative lifetimes, good agreement with experiment is also found provided the value of r_0 is taken to be 1.2. For this r_0, however, the strong coupling model is found to give equally good results. Thus for these $M4$

transitions both collective models show improvement over the simple single-particle picture.

However, Wallace (75) has shown that a refinement of the single-particle model which takes account of the readjustment of the wave functions of all the nuclear particles during the predominately one-particle transition gives very satisfactory results for the $M4$ transitions for $r_0=1.4$. If r_0 is taken to be 1.2 the agreement is even better.

VII. Appendix on Nuclear Models

a. *SM—Shell model.*—The nuclear model of Mayer & Jensen (6) in which neutrons and protons are pictured as occupying shells whose order is that prescribed by an infinitely deep well with strong spin-orbit coupling. The model does not give the exact amount of spin-orbit splitting but relies upon empirical evidence for the specification of the level order which is for particle numbers up to 64: $1s_{1/2}$, $1p_{3/2}$, $1p_{1/2}$, $1d_{5/2}$, $2s_{1/2}$, $1d_{3/2}$, $1f_{7/2}$, $2p_{3/2}$, $1f_{5/2}$, $2p_{1/2}$, $1g_{9/2}$, $2d_{5/2}$, $1g_{7/2}$.

b. *SPSM—Single-particle shell model.*—A model which pictures an odd-A nucleus as a core with zero spin plus one particle which has the ground-state spin of the shell-model state for the given number of odd particles.

c. *MPSM—Many-particle shell model.*—A model in which all particles in unfilled shells are considered to be "outside" the core. The angular momenta of these particles can combine to give ground-state or low-level spins different from the shell model values.

d. *SCCM—Strong-coupling collective model.*—This model as developed chiefly by Bohr & Mottelson (9) pictures the nucleons in unfilled shells as interacting mainly with the core. The period of the nucleon motion is considered to be short compared to that of the core so that the particles can adjust adiabatically to the core shape.

The nuclear system is described in terms of the three Eulerian angles for the coordinate axes fixed in the nucleus and two internal parameters, β and γ where β is a measure of the amount of the deformation and γ specifies the shape. In terms of β and γ the eccentricities in planes perpendicular to the three nuclear axes, when these are axes of symmetry, are:

$$e_\chi = \sqrt{15/\pi}\,\beta \sin(\gamma - \chi 2\pi/3) \qquad \chi = 1, 2, 3$$

γ is interpreted as the angle between the vector β and the Z axis for which $\chi=3$. If $\gamma=0$, the eccentricity perpendicular to the z axis is zero and perpendicular to each of the other two is $-\beta\sqrt{45/4\pi}$. The nucleus then possesses the prolate shape. If $\gamma=\pi$, the nucleus has an oblate shape. If the nucleus has no symmetry axis and alternates between prolate and oblate shape via nonsymmetrical deformations, the nuclear potential will not be a function of γ and the nucleus will be "γ-unstable" (70).

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GENERALIZED ACIDITY IN RADIOCHEMICAL SEPARATIONS[1]

By Ralph A. Horne, Charles D. Coryell

Department of Chemistry and Laboratory for Nuclear Science, Massachusetts Institute of Technology, Cambridge, Massachusetts

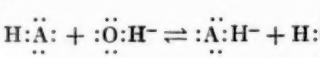
AND

Lionel S. Goldring

Radiochemistry Section, Nuclear Development Corporation of America, White Plains, New York

Introduction

Although acidic substances were recognized in classical antiquity by their acerbity and corrosiveness, the contemporary concepts of acidity are of comparatively recent genesis (1, 2, 3). The first modern definition of an acid followed immediately upon the elucidation of the Arrhenius theory of electrolytic dissociation (1884). It defined an acid as a substance which yields a proton or hydrogen ion, H^+, in aqueous solution and a base as one which yields an hydroxide ion, OH^-. This Arrhenius definition is limited to aqueous systems. In 1923 Brønsted (4, 5) and Lowry (6, 7) independently proposed a definition which avoided this restriction. They defined an acid as a proton donor, a base as a proton acceptor. A definition fundamentally different from and of greater generality than those of Arrhenius, Brønsted, and Lowry was proposed by Lewis (8, 9, 10) in 1923. He defined an acid as an electron-pair acceptor, a base as an electron-pair donor. Examples of Lewis acid-base reactions are



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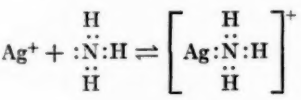

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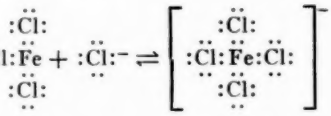

3.

The Lewis definition is more general than the Brønsted-Lowry definition and includes the whole field of complex formation, yet the concept of an acid was further generalized by Usanovich (11) in 1939. He designated as an acid any electropositive (or electrophilic) substance and as a base any nucleophilic substance. While the Arrhenius definition errs by being too specific, excluding as it does the important non-aqueous systems, on the opposite ex-

[1] The survey of literature pertaining to this review was concluded in February, 1956.

treme, the comprehensive Usanovich definition is so general as to be of meager utility although it does perform a valuable service in calling attention to the analogous relationship between acid-base and oxidation-reduction reactions.

The Brønsted-Lowry definition enables a function to be defined which quantitatively describes the acidity of a system

$$pH \equiv -\log a_{H^+} \equiv -\log (H^+)\gamma_{H^+} \qquad 4.$$

where a_{H^+} is the hydrogen ion activity, (H^+) the hydrogen ion concentration, and γ_{H^+} the activity coefficient of the hydrogen ion. The activity coefficient of a specific ion in solution is operationally meaningless (12), so $\gamma_{\pm}$, the measurable mean activity coefficient (13), is substituted for γ_{H^+}. As the concentration of an electrolyte approaches zero its activity coefficients approach unity, hence pH is effectively $-\log (H^+)$ for dilute solutions.

A useful elaboration of the Brønsted-Lowry concept of acidity has been made by Hammett (14, 15, 16) who defines a new acidity function

$$H_0 \equiv -\log a_{H^+} - \log (\gamma_B/\gamma_{BH^+}) \qquad 5.$$

or equivalently

$$H_0 \equiv pK_a - \log [(BH^+)/(B)] \qquad 6.$$

where pK_a is $-\log K_a$, K_a being the thermodynamic ionization constant of the unipositive acid form, BH^+, of the neutral base indicator, B:

$$BH^+ = H^+ + B, \quad K_a = \frac{(B)(H^+)\gamma_B\gamma_{H^+}}{(BH^+)\gamma_{BH^+}} \qquad 7.$$

An indicator is a substance whose color changes in transformation from its acid to base form. The Hammett fuction, H_0, may be interpreted as effective pH and has been particularly useful in describing the acidity of concentrated mineral acid systems.

Nearly all chemical species of interest to the nuclear chemist exhibit acid-base properties. The foregoing considerations, therefore, have many important applications to the problem of radiochemical separations where it is crucial, not only that the separation be complete or "clean," but, especially when short-lived radioactive materials are involved, that the separation techniques be relatively simple and the operations fast. Certain well-known reactions, such as the hydrolysis of metals and the precipitation of their hydroxides or sulfides, have been incorporated in traditional separation procedures. This paper will attempt to review these and more novel techniques in a brief and coherent manner and to indicate new possibilities.

Classical Acidity in Water

The acid-base properties of water set the limits of acidity in dilute aqueous solutions. The very strong acids (H_2SO_4, HCl, HBr, HI, and HNO_3) undergo proton transfer to give the hydronium ion H_3O^+, and thus their dilute water solutions contain essentially only this acid. Corrrespondingly

all very strong bases accept a proton from water to give the solvent's basic ion OH^-. The ionization of water regulates the relation between the hydrogen and hydroxide ion concentrations

$$H_2O = H^+ + OH^- \qquad K_w = (H^+)(OH^-) = 1.008 \times 10^{-14} \qquad 8.$$

Unless specifically documented, numerical constants quoted in this review are taken from the compilation of Latimer (17) which is coordinated with the U. S. Bureau of Standards report (18). It will also be convenient to refer to the negative logarithm of a constant as pK. Thus $pK_w = 14.00$.

Because of the conjugate nature of acids and bases implicit in the transfer of a proton, the ionization of the base BOH in water

$$BOH = B^+ + OH^-, \qquad K_b = \frac{(B^+)(OH^-)}{(BOH)} \qquad 9.$$

can be expressed in terms of the ionization of the conjugate acid

$$B^+ + H_2O = BOH + H^+, \qquad K_a = \frac{(BOH)(H^+)}{(B^+)} = \frac{K_w}{K_b} \qquad 10.$$

This convention will be used throughout this paper.

Any system containing a weak acid HA partially converted to its basic form A^- will have the pH value set by

$$pH = pK_a - \log \frac{(HA)}{(A^-)} \qquad 11.$$

Such a system will be relatively insensitive to pH changes on the addition of a smaller number of moles dB of strong base, and is called a buffer. The buffering power of a system can be expressed as the amount of base required to give specified shift of pH, and can be designated as $dB/d\text{pH}$

$$\frac{dB}{d\text{pH}} = 2.3mx(1 - x) \qquad 12.$$

where m is the total moles of $HA + A^-$, x is the fraction in basic form A^-. The buffering power is at a maximum of $0.58m$ when $pH = pK_a$.

Table I gives the pK_a values of a number of common acids whose properties effect buffering in inorganic reactions. Solutions of tracers must always be buffered or have the pH controlled by excess of strong acid or base to give assurance that the tracer species is in a specified acid base form.

Consider a multibasic acid H_nA, of successive K_a values K_1, K_2. . . .The fraction X_n in the form A^{-n} will follow the equation

$$X_n = \prod_1^n \frac{K_i}{(K_i + (H^+)} \qquad 13.$$

where $\prod$ denotes the multiple product. This equation shows, for instance, how the concentration $(S^=)$ of a solution of H_2S or HS^- depends on acidity. The concentration of unionized H_2S is equal $0.10\ p_{H_2S}$; the first and second pK_a values are 7.0 and 14.9. Below pH 7, the $(S^=)$ concentration is expressed as $10^{-22.9}\ p_{H_2S}/(H^+)^2$, and, between pH 7 and 14 the expression is $10^{-14.9}$

TABLE I

pK_a VALUES OF COMMON ACIDS*

Acid	pK_a	Acid	pK_a
Sulfurous H_2SO_3	1.9	Dihydrogen Phosphate Ion $H_2PO_4^-$	7.2
Hydrosulfate Ion HSO_4^-	1.9	Boric H_3BO_3	9.2
Phosphoric H_3PO_4	2.1	Ammonium Ion NH_4^+	9.3
Hydrofluoric HF†	3.2	Hydrocyanic HCN	9.4
Formic HCOOH	3.7	Hydrazinium Ion $N_2H_5^+$	8.0
Acetic CH_2COOH	4.8	Bicarbonate Ion HCO_3^-	10.4
Carbonic H_2CO_3‡	6.4	Hydrogen Peroxide H_2O_2	11.8
Hydrosulfuric H_2S	7.0	Monohydrogen Phosphate $HPO_4^=$	12
Hydrosulfite Ion HSO_3^-	7.2	Hydrosulfide Ion HS^-	(14.9)

* Reference Latimer (17); the data are roughly corrected to zero ionic strength and 25°C.

† At higher HF concentrations the reaction $HF+F^-=HF_2^-$, $K_f=3.96$ must be taken into account.

‡ All forms of dissolved CO_2 are treated as (H_2CO_3).

$(HS^-)/(H^+)$. Control of acidity can be used to regulate $(S^=)$ and thus the selective precipitation of the sulfides of different metals, since the free metal ion concentration (M) in equilibrium with precipitate M_AS_B is given as

$$(M) = K_{sp}^{1/A}/(S^=)^{B/A} \qquad 14.$$

where K_{sp} is the solubility product $(M)^A(S^=)^B$.

A detailed treatment of the common metals whose sulfides are precipitated effectively by gaseous H_2S at 100°C under varying conditions of acidity is given by Swift (19). In $9M$ HCl only As_2S_5 and As_2S_3 precipitate; in $4.5M$ H_2SO_4 to these are added HgS, CuS, Ag_2S, Sb_2S_5, Sb_2S_3, and Bi_2S_3; in $0.3M$ HCl plus $0.3M$ NH_4Cl, SnS_2, Cds, Pbs, and SnS are included; at pH = 2 (HSO_4^--$SO_4^=$ buffer), ZnS is precipitated, at pH = 6 (CH_3COOH-CH_3COO^- buffer), CoS and NiS precipitate too; and finally FeS precipitates.

Sulfide precipitation is used as one of the purification steps in many radiochemical procedures. In the collected Los Alamos Procedures (20), for instance, the elements Fe, Co, Ge, As, Ag, In, Sn, Sb, and W are specifically precipitated, and GeS_2 is used as the weighing form for this element. In many of the procedures for these and other elements sulfides are used to remove impurities at appropriate acidities. Examples include As_2S_5 in the procedure for P; As in the procedure for Ge in the presence of HF, and Ge, As, and Te in the procedure for Sb.

A very useful technique is that of sulfide scavenging to remove numerous possible impurities. In acid solution, PdS and CuS are proposed; near pH 9 (NH_4HS), TeS and SnS are used. Several elements (e.g. Mo and Re) form sulfide precipitates slowly. Bi_2S_3 precipitation speeds up the separation.

In alkaline systems some metallic sulfides dissolve to form thiosalts

$$SnS_2 + S^{=} = SnS_3^{=} \qquad 15.$$

As(V), As(III), Sb(V), Sb(III), and Sn(IV) sulfides dissolve in NH_4HS solution, Hg(II) and Sn(II) in a more alkaline NaHS-NaOH solution. Reaction No. 15 is an acid-base reaction in the Lewis sense. The problem of complex ion formation in general will be treated later in greater detail.

Metallic cations may behave as acids in the Lewis sense, water behaving as a Lewis base, thus to cite a specific example

$$Fe^{+3} + HOH = FeOH^{+2} + H^+, K_1 = (FeOH^{+2})(H^+)/(Fe^{+3}) = 1.6 \times 10^{-3} \qquad 16.$$

$$FeOH^{+2} + HOH = Fe(OH)_2^+ + H^+, K_2 = (Fe(OH)_2^+)(H^+)/(FeOH^{+2}) = 1.2 \times 10^{-3} \qquad 17.$$

$$Fe(OH)_2^+ + HOH = Fe(OH)_3 + H^+, K_3 = (H^+)/(Fe(OH)_2^+) = 9 \qquad 18.$$

where the first two constants have been taken from Hedström (21) and the solubility product of $Fe(OH)_3$ as 6×10^{-38} from Latimer (17).

Table II lists pK_1 for the first hydrolysis (Equation 16) of some common metallic cations. The appearance of (H^+) in the numerator of $K_1, K_2 \cdots K_n$ for the successive hydrolyses indicates that the extent of hydrolysis is dependent on the acidity of the system, the greater the acidity the less

TABLE II

FIRST HYDROLYSIS OF SOME METALLIC CATIONS*

Cation	pK_1	Reference	Cation	pK_1	Reference
Ce^{+4}	−0.7	(22)	Al^{+3}	4.9	(17)
Tl^{+3}	−0.7	(23)	Pb^{+2}	6.2	(17)
Hg^{+2}	0	(17)	Pu^{+4}	7.3	(22)
U^{+4}	0.7	(22)	La^{+3}	8.3	(22)
Pu^{+4}	0.7	(22)	NpO_2^+	8.9	(22)
Sn^{+2}	1.7	(24)	Zn^{+2}	9.6	(17)
Fe^{+3}	2.8	(21)	Ni^{+2}	10.7	(25, 26)
Ga^{+3}	2.6	(17)	Co^{+2}	12.3	(25, 26)
In^{+3}	3.7	(17)	Ca^{+2}	12.5	(27)
Cr^{+3}	3.8	(17)	Ba^{+2}	13.4	(27)

TWO-PROTON HYDROLYSIS TO DIMERIC PRODUCT

$2M^{+m} + H_2O = M_2O^{2m-2} + 2H^+, K_{2h}$

Cation M^{+m}	pK_{2h}	Reference
Bi^{+3}	1.2	(17)
UO_2^{+2}	6.0	(28)
Th^{+4}	7.9	(22)

* The data generally refer to zero ionic strength and 25°C.

extensive the hydrolysis. Unfortunately hydrolysis is not as simple a process as has been outlined above, polymeric products may result, as, again, in the case of iron (21, 29, 30). The first hydrolysis of some ions (Bi^{+3}, UO_2^{+2}, and Th^{+4}) is best treated as two proton-hydrolysis to a dimeric product (Table II)

$$2M^{+m} + H_2O = M_2O^{m-2} + 2H^+ \qquad 19.$$

In addition, the uncharged hydroxide is usually of very low solubility, and its ready formation may reduce the formation of intermediates, such as $Fe(OH)_2^+$. In the case of Fe(III) the acidity is approximately set by the principal reaction

$$Fe^{+3} + 3HOH \rightleftharpoons Fe(OH)_3 + 3H^+, K_T = (H^+)^3/(Fe^{+3}) = 1.7 \times 10^{-5} \qquad 20.$$

While the hydrolysis of metallic cations to hydroxides can be used to advantage to effect radiochemical separations, as, for example, in the separation procedures for Ta, Be, Se, Nb, and In (20), unanticipated hydrolysis with consequent precipitation or surface adsorption is a commonly encountered difficulty. The precipitation of ferric or lanthanum hydroxide is a familiar scavenger technique (18, 31, 32) to remove traces of radioactive contaminants which are easily hydrolyzed and sorbed on the hydrous oxide precipitate.

In radiochemical process work it is often desirable to regulate the acidity by causing partial hydrolysis of a major metallic component such as Al^{+3} to make an appreciable molarity of its first hydrolyzed state $AlOH^{+2}$. The system is called acid deficient, the degree being given as the calculated molarity of this first hydrolyzed form (33). The hydrolysis of the ion UO_2^{+2} plays an important role in radiochemistry. The first hydrolysis form UO_2OH^+ undergoes polymerization to $U_2O_5^{+2}$. Sutton (28) has determined the constants for the formation of $U_2O_5^{+2}$ and further polymer products $U_3O_8^{+2}$ and $U_3O_8OH^+$. Further discussion of the hydrolysis of the heavy metals in various valence states has been given by Kraus (22).

Complex Ion Formation

Metallic cations are, as we have seen, Lewis acids, and they have the property of forming complex compounds with Lewis bases. Such bases include anions, particularly those of weak acids, and NH_3 and its derivatives. The metallic cation generally adds successive ligands (basic groups) stepwise (34, 35) until its coordination number is reached:

$$M + A = MA, \qquad k_1 = (MA)/(M)(A) \qquad 21.$$

$$MA + A = MA_2, \qquad k_2 = (MA_2)/(MA)(A) \qquad 22.$$

. . . . . .

$$MA_{n-1} + A = MA_n, \qquad k_n = (MA_n)/(MA_{n-1})(A^-) \qquad 23.$$

or alternatively, we can speak of the over-all formation constant:

$$M + nA = MA_n \qquad \beta_n = \Pi k_n = (MA_n)/(M)(A)^n \qquad 24.$$

Total charges on the complex are obtained from the algebraic sum of +m on the metal and −a on the ligand A.

If the ligand A shows low proton affinity, like Cl^- or SCN^-, there are no pH effects in the formation of the complex. If it is NH_3 or a substituted amine or pyridine, there will be effects in the pH range where proton addition on the base competes with capture by the metal. The majority of complex-formers are the salts of weak acids H_aA, for which the fraction X_a of anion in the most basic form is given by Equation 13. Thus the gross formation constant of the complex is β_n multiplied by $(X_a)^n$, and thus strongly dependent on acidity. Prominent weak acids whose anions form useful complexes are HF, HSO_4^-, and $H_2C_2O_4$. Bjerrum (35) has given an extensive compilation of the formation constants for the inorganic complexes of a variety of metals.

In the familiar Noyes & Swift (36) scheme of qualitative analysis Sb(III) and Sn(IV) are separated from As_2S_3 as $SbCl_4^-$ and $SnCl_6^=$, Ag(I) from Hg(I) as $Ag(NH_3)_2^+$, Cu(II) and Cd(II) from Bi(III) as $Cu(NH_3)_4^{++}$ and $Cd(NH_3)_4^{++}$, and Mg(II) from Ca(II) as $Mg(C_2O_4)_2^=$.

The order of complexing strength for various inorganic anionic ligands with iron(III), for example, is $F^- > SCN^- \cong SO_4^= > HSO_4^- > Cl^- > Br^- > NO_3^- > ClO_4^-$. With silver(I), indium(III) and may other cations, however, the order for halide ions is $I^- > Br^- > Cl^- > F^-$. The stability of chlorometallic complexes varies widely from those metals which have well-known anionic complexes such as $AuCl_4^-$, $AgCl_2^-$, $PtCl_4^=$, and $PtCl_6^=$; through a number of strong ($k_1 \geqq 100$) complexing cations, BiO^+, Bi(III), Cd(II), Cu(I), Hg(II), In(III), and Tl(III); through complexes of intermediate strenth ($1 < k_1 < 100$), Ce(III), Cu(II), Fe(III), Mn(II), Th(IV), Tl(I), and U(IV); to the weak ($k_1 \leqq 1$) complexes, Cr(III), Ni(II), Pu(IV), UO_2^{++}, Zn(II) and, of course the alkali and alkali-earth metals. A number of ligands satisfy more than one coordination position on the metal cation; these are called chelates. A comprehensive review of the chelate complexes by Martell & Calvin appeared recently (37); some of the comparative inorganic chemistry of metal complexes has been discussed by Coryell (38). Typical monoacidic bidentate ligands are acetylacetone AA, and its homologues, especially trifluoracetylacetone TFA and thenoyltrifluorylacteone TTA, cupferron (nitrosophenylhydroxylamine), oxine (8-hydroxyquinoline), and dimethylglyoxime. Hydroxyorganic acids, especially polyacids like tartaric and citric acid, give polydentate anions that are good complex formers. Excellent complex formers are those of the class of ethylenediaminetetraacetic acid EDTA, with enough coordination groups to satisfy the coordination demands of most metals with one anion molecule.

Solvent Extraction

Complex formation in aqueous solution often gives rise to complexes that are appreciably soluble in organic solvents not miscible with water. This occurs readily, for instance, with some monoacidic bidentate ligands, which neutralize the charge on the cation and satisfy its coordination number (inner complexes) leading to a molecular aggregate sheathed with the organic

structure of the ligand. Examples are Ni(dimethylglyoxime)$_2$ forming above pH 5 and soluble in chloroform, Ac(TTA)$_3$ extracted by benzene above pH 5, Th(TTA)$_4$, extracted by benzene above pH 1. The use of such complexes to separate the heavy elements in various oxidation states is reviewed by Hyde (39). High specificity is achieved by pH control because of the differences in the overall formation constant β_n (Equation 24).

Our understanding of the detailed principles of the solvent extraction of inorganic complexes is imperfect indeed. Extraction of halide complexes of multivalent cations from HCl, HBr, or HI solution is used frequently. The species taken up by the organic layer is usually HMX_4 for trivalent ions: Au, Tl, Ga, In, and Fe, and neutral MX_4 for tetravalent species like Te and Ge. The extraction properties depend somewhat on H^+ for the trivalent elements because of participation of the proton in the extracted molecule, and on H^+ for the tetravalent species because of the necessity to suppress hydrolysis, but the main specificity comes from control of halide ion concentration. The extraction coefficient E (ratio of total metal concentration in the organic phase to that in the aqueous phase) is regulated appreciably by acidity in the organic phase, which is set by ionization constants and concentrations of the different acids extractable from the water phase, as outlined by Saldick (40). Schonken & Irvine (41) on the basis of studies of the extraction of $HAuCl_4$ into various solvents have concluded that, in addition to a pronounced dielectric constant dependence, the extraction coefficient increases as the Lewis basicity of the solvent increases. Other important factors in solvent extraction are the acid strengths of the species in the organic phase, solvation of the species, and polymerization (42, 43).

Some neutral nitrates prove to be fairly extractable into organic solvents, for example, $UO_2(NO_3)_2$ into diethyl ether. Acidity must be controlled in this extraction to prevent hydrolysis in the water phase. Its efficiency is much higher from a concentrated nitrate solution of salting agents (HNO_3, NH_4NO_3, $Ca(NO_3)_2$, or $Al(NO_3)_3$), providing common ion. The extraction is more effective with tetranitrates of Th and Pu. At high acidities, tracer Pa is also extracted (39). Rather highly oxygenated solvents such as methyl isobutyl ketone, mesityl oxide, and dibutoxytetraethylene glycol (pentaether) are also effective with HNO_3 and nitrate salts for heavy metal extraction (39).

A recently developed technique for solvent extraction of nitrates is based on solutions of tributylphosphate (TBP) in inert hydrocarbon, with salting agents in the aqueous phase. The chief species extracted are $UO_2(NO_3)_2$, and the nitrates of Ce(IV), Zr(IV), Hf(IV), Pu(IV), and U(IV). The use of TBP in heavy element separations is discussed by Hyde (39), and conditions for minimizing fission-product contamination (lower acidity) are discussed by Bruce (44).

Ion-Exchange Separations

Consider the following acid-base reaction

$$M^{+m} + mHZ \rightleftharpoons mH^+ + MZ_m \qquad 25.$$

where the acid HZ is a cation exchange resin, that is to say, Z, rather than being a mobile anion, is a function group attached to a fixed resinous matrix. Strongly acidic cation exchange resins consist commonly of sulfonic acid functional groups attached to a polystyrene matrix; in weakly acid resins the function group may be a carboxylic group or a phenolic hydroxide. While the weak resins preferentially hold hydrogen ion by a large factor, the strong cation exchange resins hold hydrogen ion, ammonium ion, and the alkali metal ions about equally firmly, the order of decreasing bonding being $Cs^+ > Rb^+ > K^+ > NH_4^+ > Na^+ > Li^+ > H^+$. Cations are held more firmly the more positive their charge (45). Thus by complexing a metallic cation with an anionic ligand, and thereby decreasing its positive charge, the specificity of the metal's cation-exchange resin absorption can be varied at will, and separations, often exceedingly difficult by any other technique, readily effected. When the ligand is an anion of a weak acid the separation will be dependent on the pH.

Perhaps the best known application of ion-exchange resin techniques is to the separation of the rare earths, a separation which, because of the similarity of their chemistry, was of particular difficulty, frequently requiring recourse to such an extremely arduous and tedious technique as fractional crystallization. Rare earths and actinide earths are now separated with facility by selective elution of their citrate complexes from a cation exchange resin packed column (46) thus achieving the equivalent of many successive operations. Lactic acid has also been used as a complexing agent to separate rare-earth fission products by cation exchange (47), advantageous uza-tili tion of the pH sensitivity of actinide elution with ammonium α-hydroxy-isobytyrate has been reported (48), and chloride ion has been used to complex and separate the actinide group from the lanthanide group with a cation exchange resin (49) and thiocyanate ion to separate the actinides and lanthanides with both cation and anion exchange resins (50). Data for the absorption of metallic cationic species on cation-exchange resions may also be analyzed to yield the successive formation constants of metallic complex ions (51). We might also note in passing, as evidence for the acidic character of cation exchange resins in the hydrogen ion form, their catalysis of certain acid-catalyzed organic reactions (52, 53).

In the case of anion-exchange resins, although some features are apparently analogous to the corresponding case of cation exchangers, many are not, and the situation is presumably complex and the state of our knowledge imperfect. The functional group in these resins is a quaternary amine. The affinity of the strong anion-exchange resin for hydroxide is weak, being about 0.1 that for chloride ion (54), but can be increased by the substitution of an ethanol group on the quaternary amine. The hydroxide form of the strongly basic anion exchange resins is sometimes used to precipitate trace amounts of metals as hydroxides, for example Y^{90} or La^{140} from Sr^{90}-Y^{90} or Ba^{140}-La^{140} mixtures (55).

Anion-exchange resins often show a very high affinity for metal complexes, and this selectivity can be exploited for numerous radiochemical separations,

as shown particularly in the extensive work of Kraus and coworkers reviewed by Kraus & Nelson (56). If we plot the logarithm of the distribution coefficient, D (the concentration of metal in the resin divided by the concentration of metal in solution), *versus* the concentration of complexing anion in solution, the curve rises to a maximum, with log D sometimes as great as six or even larger, and then falls. Kraus, Nelson & Smith (57) pointed out certain similarities between the solvent extraction and anion exchange behavior of metal halides. It is also found (56) that substitution of LiCl for HCl leads to an increase of D for many metals in concentrated chloride solution.

On the basis of concepts elucidated by Fronaeus (58), Coryell & Marcus (59) attribute the maxima in log D to the markedly preferential absorption of the neutral complex MA_m from the aqueous solution into the resin phase:

$$RA + MA_m = RMA_{m+1} \qquad 26.$$

The position of the maxima is related to the strength of the metal complexes, while the strength of absorption of the resin, as measured by $D_{maximum}$, now appears to be related to the Lewis acidity of the neutral complex as measured by the formation constant, k_{m+1}, of MA_{m+1}^- (60). The fall in the absorption curve may be attributed to the formation in the aqueous phase of higher complexes, MA_{m+1}^-, $MA_{m+2}^=$, and perhaps acids such as HMA_{m+1}. The LiCl effect (56) may be interpreted as evidence for the importance of formation of unionized acids like HMA_{m+1}, but it may be also be explained by great changes in activity coefficients in the resin phase and in the aqueous solution.

The use of chloride ion as a complexing ligand in anion-exchange resin separations has received considerable attention and this work was reviewed recently by Kraus & Nelson (56). Examples of such separations are Fe(III)—Pa(V) (57) and Nb(V)—Ta(V) (61) from HCl-HF mixtures of varying composition; Ni(II), Mn(II), Co(II), Cu(II), Fe(II), and Zn(II) from solutions of varying HCl concentration (62); Co(II), Cu(II), Zn(II), and Ga(III) from HBr solutions (63); and the use of organic complexing agents such as citrate ion and EDTA for alkaline-earth and rare-earth anion-exchange resin separations (56). For further information on the separative and analytical uses of ion-exchange resin techniques the reader is referred to the text by Samuelson (64) and the reviews by Finston & Miskel (55) and Schindewolf (65).

Generalized Acidity

The empirical justification of the Hammett acidity function (Equation 6) lies in the fact that this function appears to be independent of the nature of the indicator used for its measurement and a property of the solution only; its utility in that it avoids, by virtue of being an experimentally measured quantity, the ambiguities involved with the changing activity coefficients at larger concentrations while at the same time becoming equivalent to pH in

dilute solutions. Furthermore the concept is extendible to very strongly acidic and non-aqueous systems. Figure 1, taken from the recent detailed review of Paul & Long (66), shows the variation of the acidity function, H_0, with the acid molarity of aqueous solutions of several common mineral acids. This figure shows that $HClO_4$ and H_2SO_4 are among the strongest acids whereas HCl and HNO_3 are noticeably weaker.

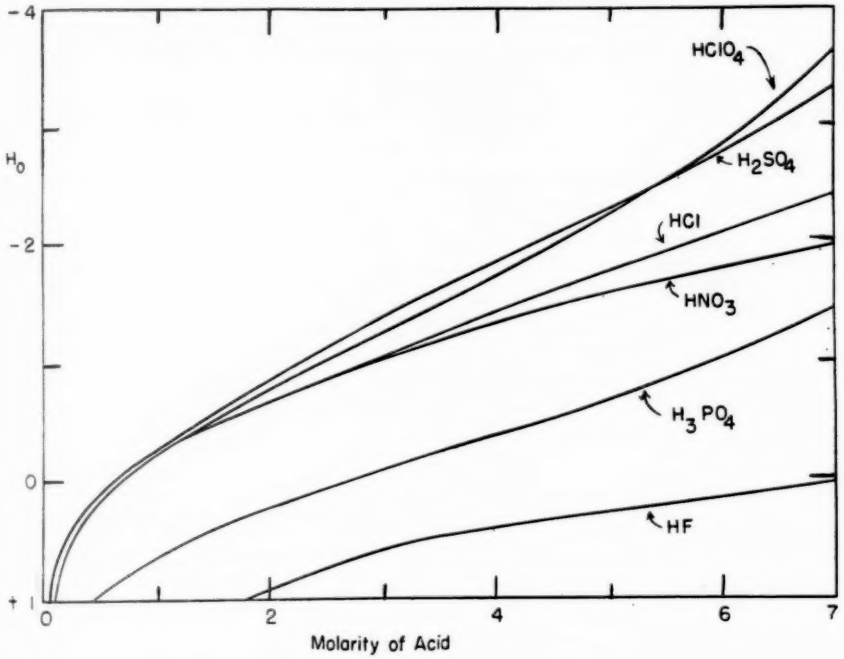

FIG. 1. The Hammett Acidity Function H_0 for some mineral acids as a function of molarity. Reproduced with permission from Paul & Long (66).

The H_0 values for H_2SO_5 systems have received considerable attention (67 to 71), having been studied up to 100 per cent H_2SO_4 and on into the H_2SO_4-SO_3 region. For this system the empirical relation

$$-H_0 = 0.5M - 0.3 \qquad 27.$$

where M is the molar concentration, is valid in the region five per cent ($0.5M$) to 95 per cent ($18M$). The curve then rises steeply to $H_0 = -11.10$ at 100 per cent H_2SO_4 and levels off towards $H_0 = -12.6$ at 105 per cent H_2SO_4 (22.3 per cent SO_3-H_2SO_4). Lewis & Bigeleisen (67) extended the scale, using the vapor pressure of SO_3, to obtain a limiting value of $H_0 = -14.6$ for pure SO_3.

The foregoing data indicate the enormous effective acidity of con-

centrated H_2SO_4, pure H_2SO_4 having a proton activity of some 10^{11} times greater than $1M$ H_2SO_4, and thus explain its great solvent power. The solubility of CrO_3 in concentrated H_2SO_4 to form $HCrO_3^+$ and CrO_2^{+2} ions suggests its basic properties, similarly for the pentoxides of Nb(V) and Ta(V), which also dissolve to form cationic species in this solvent. On the other hand the distillation RuO_4 and Tc_2O_7 from fission product mixtures from concentrated $HClO_4$ or H_2SO_4 solution indicates the low basicity of these oxides.

Concentrated acids are often used as catalysts to equilibrate aromatically bound deuterium and tritium with water for their isotopic determination. Gold & Satchell (72) have established that the logarithm of the rate of such exchange in several mineral acids is a linear function of $-H_0$. Many other acid catalyzed reactions can be correlated with H_0 or related functions (such as J_0 which contains a term for the activity of water) (73).

Hammett acidity functions for common mineral acids in a number of nonaqueous and mixed solvent systems are reviewed by Paul & Long (66).

The Hammett function, H_0, is, as the subscript indicates, defined in terms of an indicator of zero charge in its base form. Alternative scales for other charge classes of indicators, H_{+2}, H_+, H_-, etc., have been set up (74 to 77). It has been shown (66, 71) that these functions when plotted *versus* M of H_2SO_4 differ by constants indpendent of M above about $4M$; thus intercomparisons can be made.

In addition to acidic systems, the Hammett acidity function has also been evaluated for strongly basic media. Deno (77) has estimated that the H_0 of 90 per cent N_2H_4—10 per cent H_2O is plus 19, Schwarzenbach & Sulzberger (78) that a saturated aqueous NaOH solution is plus 19, and Conant & Wheland (79) have measured basicities in ethereal solution that correspond to H_0 values of about plus 31.

Non-Aqueous Solvents and Liquid Salt Systems

Proposals have been made by Starke (80) for exploiting acid-base reactions in non-aqueous media for radiochemical separations, in particular with the fused chloride salts of substituted amines.

An example of acidity in a non-aqueous solvent is given by processes which have been developed for the dissolution of uranium metal in interhalogen compounds such as BrF_3, BrF_5, and ClF_3 (81). In the halogen fluoride systems the loss of a fluoride ion is equivalent to the gain of a proton in aqueous systems, i.e.,

$$2BrF_3 \rightleftharpoons BrF_2^+ + BrF_4^- \qquad 28.$$

The pure interhalogen compounds BrF_3 and BrF_5 are ionized sufficiently to dissolve uranium metal at an appreciable rate. Increase in the reaction rate with addition of bromine indicates that Br^+ is a stronger acid than BrF_2^+ which has in turn been found to be a stronger acid than BrF_4^+. Addition of a salt which can act as a fluoride ion acceptor, such as SbF_5, spectacularly

accelerates dissolution rate. Uranium reaction products also accelerate solution rate; this is explainable in terms of acidity by assuming the formation UF_5^- of $UF_6^=$.

Classical metallurgy, ceramics, and glass manufacture can, to a major extent, be considered to involve the chemistry of liquid salt systems and thus to show many instances of acid-base reactions. For many fused salt systems and systems of oxides in particular, the concept of acidity advanced by Lux [(82), cf. also (83)] is often useful. Analogous to the Brønsted-Lowry definition of acidity

$$\text{Acid} \rightleftharpoons \text{Base} + H^+ \qquad 29.$$

he defined an acid in oxide systems by the relation

$$\text{Base} \rightleftharpoons \text{Acid} + O^= \qquad 30.$$

Recently methods have been developed for the processing of nuclear reactor fuels which use liquid-salt media. The most prominent of these is the continuous processing of a liquid reactor fuel by means of fused salts, an area of chemistry referred to as pyrochemistry. Of the few systems that have been discussed publicly, attention is directed to the work for the bismuth liquid-metal fuel reactor (LMFR) (84).

In the proposed operation of this system all the fission elements of groups IA, IIA, and IIIA can be oxidized and removed into a liquid salt phase (KCl-LiCl eutectic) using $MgCl_2$ as oxidant.

In the Usanovich sense, an oxidant (such as $MgCl_2$) is an acid and the free energies of the reactions such as

$$3MgCl_2 \text{ (salt phase)} + 2La \text{ (Bi phase)} = 3Mg \text{ (Bi phase)} + 2LaCl_3 \text{ (salt phase)} \qquad 31.$$

can be related to acidities of component acids LiCl, $MgCl_2$, $LaCl_3$, and UCl_3. The effective acidity may be controlled (buffered) by regulating the amounts of Mg and $MgCl_2$. The fission elements are in small concentrations compared to uranium and bismuth so for all practical purposes the buffer concentrations remain constant.

Another application of fused salts to reactor fuel processing has been in the dissolution of alloyed fuel elements, e.g., Zr-U alloy, for subsequent processing by distillation of UF_6. A number of alloys are not dissolved by BrF_3 at an appreciable rate. However, in liquid salts (50 mole per cent NaF, balance ZrF_4) at an elevated temperature dissolution can be readily achieved by BrF_3. Since BrF_3 is quite expensive, it is desirable to replace it by a cheaper acid and dissolution can be achieved quite readily in the fused salts by bubbling HF through the mixture. This results in the formation of UF_4 and ZrF_4. Subsequently, the uranium can be readily separated by fluorinating it to UF_6 by F_2, BrF_3 or BrF_5.

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THE MASSES OF LIGHT NUCLIDES[1]

By J. Mattauch, L. Waldmann, R. Bieri,[2] and F. Everling
Max-Planck-Institut für Chemie, Mainz, Germany

1. Introduction

One is interested in the masses of nuclides and their errors mainly for two reasons: (*a*) The system of masses represents a brief expression of the experimental material from which they have been calculated. (*b*) From the masses can be calculated a series of binding energies, of substantial interest in nuclear physics. Thus, the binding energies $E(N, Z)$, the capture energies of the last nucleon or pair of nucleons, can be computed, as well as the energy differences of neighboring isobars, corresponding approximately to the first derivatives of the energy surface $E=E(N, Z)$ in the direction of N and Z. The pairing energies, which may be interpreted as second derivatives of E with respect to N and Z, respectively, can also be calculated.

The experimental data used for the calculation of masses and which we shall call Q-values for short have been compiled in a masterly way in a series of reports which appeared in the October issue 1954 of the *Review of Modern Physics*. For the calculation of masses of the light nuclides data may be used in the articles by Van Patter & Whaling (1) on nuclear disintegration energies, by King (2) on total β-disintegration energies, and by Duckworth *et al.* (3) on mass spectroscopic doublet values.

The connection between the experimental data (Q-values) and the masses or the binding energies will be discussed in the next section. Now, in the region of light nuclides the number N of Q-values is considerably larger than the number n of masses to be calculated from the former. In other words, the problem is heavily overdetermined and therefore is a typical problem of least squares adjustment, similar to the evaluation of physical constants carried out since 1929 from time to time in a series of model papers (4) especially by R. T. Birge, by J. W. M. DuMond and by others. A list of formulae of least squares adjustment required for our purpose is made up in Section 3. As far as we know, the first somewhat extensive least squares adjustment of this kind has been carried out by the two senior authors for the masses of the Isotopic Report 1949 (5). Among later similar least squares adjustments using newer data we call to notice especially the papers by Li *et al.* (6), by Drummond (7), and by Wapstra (8). The N given Q-values, which are to be used for the evaluation of n masses, have to satisfy $N-n=r$ conditional relations (cycles) which have to be linearly independent of each other. The setting up and the discussion of these conditions which is done in section 4 renders it possible, as shown in section 5 to debar from the least squares calculation those experimental data which are in disagreement with

[1] The survey of literature pertaining to this review was concluded in March, 1956.

[2] Now at the Physics Department, University of Minnesota, Minneapolis, Minnesota.

the balance of the remaining data and which would therefore falsify the adjusted values were they used as input values. Furthermore, this procedure is useful as a first approximation to the least squares adjustment.

2. The Connection Between Q-Values and Masses or Binding Energies, Respectively

The mass M of an atom built up of N neutrons, Z protons, and Z electrons is connected with the binding energy E by the well known equation

$$M = N^1n + Z^1H - E \qquad 1.$$

Here, M, ^1H, and ^1n denote the total rest masses of the corresponding atoms. However, equation 1 evidently remains unchanged, if by M, etc., the mass excesses $M = M_{\text{total}} - (N+Z)\frac{1}{16}{}^{16}\text{O}$, etc. are understood, which are defined by the mass unit (1 mass unit $= \frac{1}{16}{}^{16}\text{O}$), agreed upon by convention. In this paper mass excesses are meant always whenever masses are referred to directly. They may be stated *ad libitum* in mass units (practically always in mMU) or in energy units (Mev) which is expressed already in equation 1 except for the factor c^2. To use total masses is unnecessary for purposes of nuclear physics and would be rather cumbersome especially if stated in energy units.

To specify a nuclide we employ for our purposes, instead of N and Z, the mass number or number of nucleons $A = N+Z$ and the neutron excess $I = N-Z = 2 \cdot T_\zeta$ (T_ζ = isotopic spin). Therefore $N = \frac{1}{2}A + T_\zeta$, $Z = \frac{1}{2}A - T_\zeta$ and

$$M(A, T_\zeta) = \tfrac{1}{2}A(^1n + {}^1\text{H}) + T_\zeta(^1n - {}^1\text{H}) - E. \qquad 2.$$

The experimental data at our disposal for the evaluation of masses, the binding energies to be calculated from the masses, as well as other interesting quantities are always very simple linear combinations of a few masses distinguished by the property that the mass numbers cancel out. In these combinations the masses are multiplied by simple numbers κ, integers or fractions, which in analogy to chemical reactions we shall denote as "stoichiometric coefficients." Such a linear combination we shall call a Q-value. It is, therefore,

$$\sum_i \kappa_i M_i = Q, \quad \text{if} \quad \sum_i \kappa_i A_i = 0. \qquad 3.$$

We discern three kinds of experimentally measured Q-values used for the calculation of light masses: Q_r, Q_β and Q_m.

1) *Energies Q_r liberated by nuclear reactions.*—For example, the energy liberated by the reaction $^{14}\text{N}(\alpha, p)^{17}\text{O}$ is $Q_r = {}^{14}\text{N} + {}^4\text{He} - {}^1\text{H} - {}^{17}\text{O}$. There are usually four and, with photodisintegrations, three stoichiometric coefficients κ_i differing from zero and these are mostly $+1$ or -1. One of the few exceptions is the reaction $^6\text{Li}(d, \alpha)^4\text{He}$ where $Q_r = {}^6\text{Li} + {}^2\text{D} - 2\ {}^4\text{He}$. Since by nuclear reactions no conversion of neutrons into protons or vice versa is taking place, so that T_ζ is not changed, it is always

$$Q_r = -\sum \kappa_i E_i, \quad \text{since} \quad \sum_i \kappa_i T_{\zeta i} = \Delta T_\zeta = 0 \qquad 4.$$

as follows from equation 2 and 3. From Q_r-values the binding energies of all nuclei reached by nuclear reactions may be computed. The Q_r-value of the most simple reaction, the capture of a neutron by a proton, yields the binding energy of the deuteron: $^1\mathrm{H}(n, \gamma)^2\mathrm{D} = E_d$. This and the Q_r-values of the (n, γ), (p, γ) and (d, γ) reactions using ^{2}D as a target yield easily the binding energies E_t, E_τ and E_α of the other particles used as nuclear projectiles, and so on.

The agreement upon a standard, necessary for the calculation of masses, e.g., $^{16}\mathrm{O} = 0$ means according to equation 2, that

$$(^1n + {}^1\mathrm{H}) = \tfrac{1}{8}E(^{16}\mathrm{O}). \qquad 5.$$

Since Q_r-values yield no information about $(^1n - {}^1H)$ only masses of nuclides with $T_\xi = 0$ can be computed from them:

$$M(A, T_\xi = 0) = \frac{A}{16}E(^{16}\mathrm{O}) - E(M). \qquad 6.$$

Quite generally one can say that from Q_r-values only the masses of such nuclides can be computed which in the $A - T_\xi$ diagram lie on the straight line defined by the origin and the standard.

2) *Total β-disintegration energies* Q_β.—Here the linear combination is reduced to a difference between the masses of the β-active nuclide and of the end product. Q_{β^+} denotes as is usual, the disintegration energy increased by twice the rest mass of the electron. It is, therefore,

$$Q_{\beta\pm} = M_a - M_e = \mp\,(^1n - {}^1\mathrm{H}) - (E_a - E_e),$$
$$\text{since} \quad \Delta T_\xi = \mp 1 \quad \text{for} \quad \beta^\pm \text{ decay} \qquad 7.$$

Q_β-values alone yield of course only the masses of nuclides which are isobars of the standard. However, they comprise the important neutron-proton-difference and together with Q_r-values all masses may now be computed according to equations 2 and 4.

To be sure, the statement just made is valid only if one assumes that the rest mass m_ν of the neutrino vanishes, an assumption which is always made in mass-calculations of this sort. This assumption is supported by measurements near the maximum energy of β-spectra using Fermi's theory of β-decay or one of its variants (9). If $m_\nu \neq 0$, it would have to be added to the left side of equation 7. Essentially, therefore, Q_β-values of β^+ and of β^--disintegrations would enable us to compute $(n^1 - \mathrm{H}^1)$ as well as m_ν, since

$$(n^1 - {}^1\mathrm{H}) + m_\nu = -Q_{\beta^+} - (E_a - E_e)$$
$$(n^1 - {}^1\mathrm{H}) - m_\nu = Q_{\beta^-} + (E_a - E_e). \qquad 8.$$

However, as the β^+-disintegration energies most precisely measured are stated with an error of at least ± 5 kev, the result would be of little use, even if we could neglect the errors of the corresponding binding energies, since one knows that the rest mass of the neutrino is certainly smaller than 5 kev. If besides Q_r-values only Q_{β^-}-values were used for the calculation of masses or if the Q_{β^-}-values entered into the computation with a much higher weight than the Q_{β^+}-values (as is the case in our least squares adjustment),

then all masses would have to be enlarged by $T_\zeta \cdot m_\nu$. This follows from equation 2 and the second equation 8, if afterwards by other experiments a value of m_ν differing from zero were established.

3) *Mass spectroscopic doublet values* Q_m.—Here

$$Q_m = ({}^1n - {}^1\mathrm{H})\Delta T_\zeta - \sum_i \kappa_i E_i, \qquad 9.$$

where ΔT_ζ may assume diverse values. Q_m-values yield the masses of stable and even of unstable nuclides, as far as the latter may be obtained in amounts sufficient for measurement, with the exception of ${}^1n$. Therefore no binding energies E can be computed from Q_m-values. Only with the help of at least one or more Q_r-values yielding the $\sum_i \kappa_i$ of equation 9 one can compute, $({}^1n - {}^1\mathrm{H})$ and with it the binding energies of nuclei, e.g., $({}^1n - {}^1\mathrm{H}) = E_d - ({}^1H_2 - {}^2D)$. Together with a very precisely measured Q_β-value, e.g., that of ${}^3\mathrm{T}(\beta^-){}^3\mathrm{He}$, the corresponding Q_m-value $({}^3\mathrm{T} - {}^3\mathrm{He})$ would yield the neutrino mass or an upper limit of the neutrino mass.

From the masses obtained by the least squares adjustment the binding energies $E(N, Z)$ may be computed by solving equations 1 or 2. All the other binding energies mentioned in the introduction which are linear combinations of masses, i.e., Q-values in the sense of equation 3, may also be computed.

3. General Procedure of Least Squares Adjustment (10)

There may be given N experimentally measured values with their errors $(Q_I \pm q_I,\ I = 1, 2, \cdots, N)$ from which $n < N$ masses $(M_i,\ i = 1, 2, \cdots, n)$ with their errors are to be calculated. The list of adjusted values $Q_I^* \pm q_I^*$ has to be set up in such a way that contradictions disappear, i.e., that each way of computation leads to the same adjusted mass excesses $M_i^* \pm m_i^*$. The absolute values of the deviations $v_I = Q_I - Q_I^*$ have to turn out the smaller, the more precise Q_I is measured, i.e., the smaller the error q_I is stated. According to Gauss this is best accomplished by demanding that

$$\sum_I p_I v_I^2 = C \sum_I (Q_I - Q_I^*)^2 q_I^{-2} = \text{Minimum}. \qquad 10.$$

Here we have introduced[3] for each measured value the weight $p_I = Cq_I^{-2}$.

a) The method of indirect observations.—One is always dealing with indirect observations always when the quantities to be determined (here the masses) are not accessible to direct measurement but, on the contrary, have to be computed from other measurable quantities (here the Q-values). According to equation 3 we have therefore N equations

$$Q_I^* = \sum_i \kappa_{Ii} M_i^* (i = 1, 2, \cdots, n;\ I = 1, 2, \cdots, N), \qquad 11.$$

[3] In the simple case where the Q_I are measurements of one and the same quantity Gauss's condition (see equation 10) yields as adjusted, for the most plausible value Q^* the weighted or the arithmetic mean depending upon whether the weights p_I of the diverse measurements Q_I differ from each other or are all equal. For physicists this certainly is one of the reasons for preferring Gauss's method of least squares to all other proposed methods.

where the κ_{Ii}'s are known coefficients (the "stoichiometric coefficients" of nuclear reactions). It is supposed that an approximate value $M_i^{(0)}$ of each mass is known. With the help of the abbreviation

$$Q_I^{(0)} = \sum_i \kappa_{Ii} M_i^{(0)} \qquad 12.$$

equation 11 may be written

$$Q_I^{(0)} - Q_I^* = \sum_i \kappa_{Ii}(M_i^{(0)} - M_i^*). \qquad 11a.$$

As is well known, Gauss's condition (equation 10) leads then to the n linear normal equations

$$\sum_k a_{ik}(M_k^{(0)} - M_k^*) = \lambda_i, \qquad 13.$$

where

$$a_{ik} = \sum_I \kappa_{Ii}\kappa_{Ik} q_I^{-2}, \quad \lambda_i = \sum_I \kappa_{Ii}(Q_I^{(0)} - Q_I) q_I^{-2} \qquad 14.$$

From equation 13 the adjusted masses M_i^* are to be computed. Denoting as $(a^{-1}{}_{ik})$ the inverse matrix of (a_{ik}) one obtains from equation 13.

$$M_i^* = M_i^{(0)} - \sum_k a^{-1}{}_{ik}\lambda_k. \qquad 15.$$

Substitution of the values M_i^* into equations 11 or 11a yields then the adjusted values Q_I^* of the input data. Note that the matrix (a_{ik}) is positive-definite, since it descends from a sum of squares, cf. equation 10. Therefore, in particular, $a_{ii} > 0$, which is also seen directly from equation 14.

The matrix $(a^{-1}{}_{ik})$ permits at the same time the computation of the adjusted error q^* of any linear function

$$Q^* = \sum_i \kappa_i M_i^* \qquad 16.$$

of the masses. Replacing M_i^* according to equations 15 and 14 by the Q_I and using then Gauss's law of propagation of errors one obtains

$$q^{*2} = \sum_I \Big(\sum_{i,k} \kappa_i a^{-1}{}_{ik}\kappa_{Ik} q_I^{-2}\Big)^2 q_I^2 = \sum_{i,k} a^{-1}{}_{ik}\kappa_i\kappa_k. \qquad 17.$$

The adjusted error of the masses M_i^* is, therefore, given by

$$m_i^{*2} = a^{-1}{}_{ii}. \qquad 18.$$

The matrix $(a^{-1}{}_{ik})$ is also positive-definite just as is the matrix (a_{ik}); this must be so, because each q^{*2} calculated according to equation 17 has to be positive. Instead of $(a^{-1}{}_{ik})$ one may also use the dimensionless correlation matrix of

$$c_{ik} = a^{-1}{}_{ik}\ (a^{-1}{}_{ii}a^{-1}{}_{kk})^{1/2}, \quad c_{ii} = 1 \qquad 19.$$

and one has, instead of equation 17,

$$q^{*2} = \sum_{i,k} c_{ik}\kappa_i\kappa_k m_i^* m_k^*. \qquad 20.$$

A note is still to be made about the approximate values $M_i^{(0)}$. Suppose the measured input values Q_I are arranged according to increasing errors q_I and select from the equation 11 in this sequence just n linearly independent equations, which then permit an unambiguous calculation of the

masses from the Q-values. These masses may then be used as approximate values $M_i^{(0)}$. This selection has the effect that, at the final least squares calculation of the values M_i^*, one can manage with the least possible accuracy of the a^{-1}_{ik}, since then according to equation 14 the λ_i-values turn out to be relatively small.

b) The method of conditional observations.—To render the next section more intelligible we shall consider briefly this second method of least squares adjustment. Instead of computing the values M_i^* immediately, one may also begin with adjusting the values Q_i only, starting from the observation that between them r linearly independent equations

$$\sum_I \alpha_{\rho I} Q_I^* = 0 \; (\rho = 1, 2, \cdots, r; r = N - n) \qquad 21.$$

have to be fulfilled, which result from the equations 11, by elimination of the M_i^*. The experimental values Q_I will not satisfy equations 21 exactly, but will give the residuals ϵ_ρ:

$$\sum_I \alpha_{\rho I} Q_I = \epsilon_\rho. \qquad 22.$$

Gauss's condition (equation 10), and the conditions (equation 21) are satisfied by

$$Q_I^* = Q_I - q_I^2 \sum_\rho k_\rho \alpha_{\rho I}, \qquad 23.$$

whereby Lagrange's parameters ("correlates") k_ρ are obtained by

$$\sum_\sigma b_{\rho\sigma} k_\sigma = \epsilon_\rho \quad \text{or} \quad k_\rho = \sum_\sigma b^{-1}_{\rho\sigma} \epsilon_\sigma. \qquad 24.$$

Here we have used the abbreviation

$$b_{\rho\sigma} = \sum_I \alpha_{\rho I} \alpha_{\sigma I} q_I^2, \qquad 25.$$

$(b^{-1}_{\rho\sigma})$ denotes the inverse matrix of $(b_{\rho\sigma})$. However, the least squares calculation according to this method turns out to be simpler than according to the method a) only if many values Q_I are given which are connected by only a few equations of condition ($r \ll n$); for this reason we do not follow up this method any further here.

Finally we note that, supposing the true errors of the input data Q_I follow a Gaussian distribution, the value of the minimum of equation 10 ought to turn out:

$$\sum_I p_I v_I^2 = (N - n)C = rC. \qquad 26.$$

For the r.m.s. error of an observation of unit weight before the least squares adjustment R_i (internal consistency) and after the least squares adjustment R_a (external consistency) respectively are given (11) by

$$R_i^2 = C / \sum_I p_I \quad \text{and} \quad R_a^2 = \sum_I p_I v_I^2 / \left[r \sum_I p_I \right] \qquad 27.$$

And the expectation value for the ratio of both errors, in the case that q_I are meant to be r.m.s. (and not probable) errors, is

$$R_e/R_i = \left[\sum_I p_I v_I^2/rC\right]^{1/2} = 1; \qquad 28.$$

this value is expected with a relative error of $\pm(1/2r)^{1/2}$. If the q_I are meant to be the probable errors of the input data Q_I, as is frequently stated in American literature, then equation 28 would have to be replaced by

$$R_e/R_i = 0.6745\left[\sum_I p_I v_I^2/rC\right]^{1/2} = 1. \qquad 28a.$$

After having carried out the least squares adjustment these relations render possible a test whether the errors q_I are assumed correctly.

In an analogous way each conditional relation 22 can be tested individually by its ratio $l_\rho = R_e/R_i$. Because if there is only one—the ρth—condition, one obtains, since $r=1$

$$l_\rho = \left[\sum_I p_I v_I^2/C\right]^{1/2} = \left[\sum_I v_I^2 q_I^{-2}\right]^{1/2} = \epsilon_\rho/\sqrt{b_{\rho\rho}}, \qquad 29.$$

where the values of

$$v_I = Q_I - Q_I^* = q_I^2\alpha_{\rho I}\epsilon_\rho/b_{\rho\rho} \quad \text{and} \quad \sum_I \alpha_{\rho I}^2 q_I^2 = b_{\rho\rho} \qquad 30.$$

are substituted according to equations 23, 24, and 25, specialized for a single one—the ρth—condition.

4. The Setting Up of the System of Reaction Cycles

In the next section we have to examine which of the experimental data are to be admitted as input data for the least squares adjustment. This sifting of the Q-values is best done by setting up the r conditional relations 22 and by answering the question which maximum l_ρ-value ought to be admitted. The conditions 22 may be obtained easily in the following three ways:

(a) By going around a closed polygon in Figure 1 and adding up thereby the Q_τ- and Q_β-values of successive reactions and β-decays with the right sign (positive by going in the direction of the arrow and negative by going in the opposite direction), the binding energies of the nuclei building the corners of the polygon cancel out. Using as auxiliary quantities with nuclear reactions the binding energies E_d, E_t, E_τ, and E_α of the projectiles and with β-decays the neutron-proton-difference ($^1n - {}^1\mathrm{H}$), and adding them to the sum above with the right sign given by equation 4 and equation 7, one obtains each time a linear combination of the conditional relations 22. Table I gives a series of examples of such conditions known as "reaction cycles." The processes denote thereby in an easily understandable way the values Q_I to be inserted here from Table III; the auxiliary quantities compiled in Table II are to be added. We shall return to the latter in more detail later on as well as to the procedure used in setting up the system of cycles. Here we note that the coefficients $\alpha_{\rho I}$ are always $+1$ or -1 with the exception of the auxiliary quantities.

(b) From Q_m-values one can also easily set up similar cycles, if the number

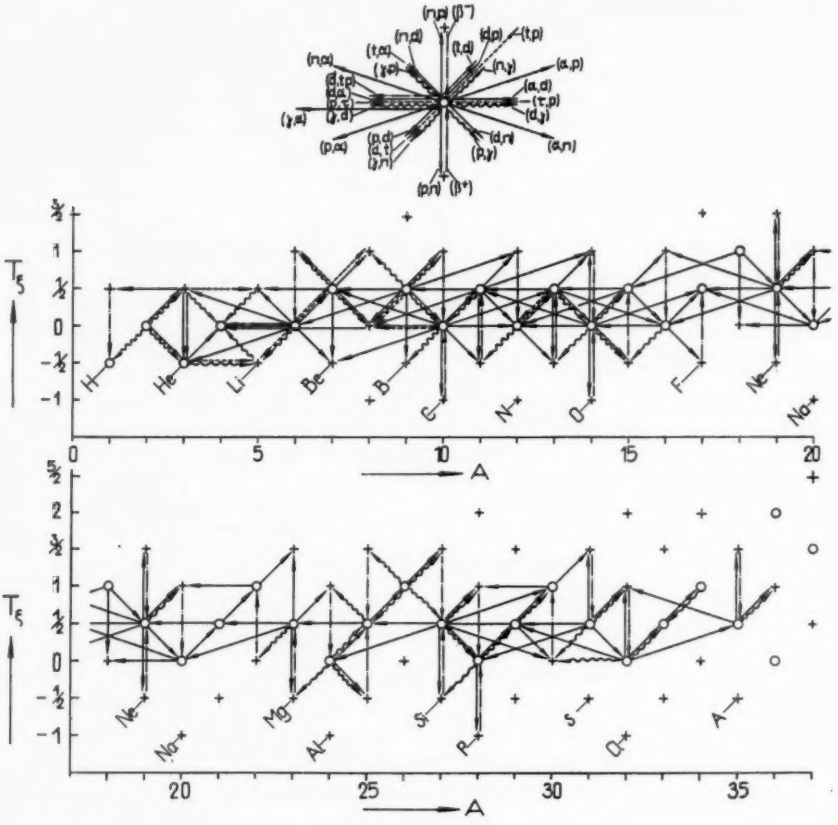


FIG. 1. $A-T_\zeta$—Diagram of ($n=70$) nuclides showing ($N=196$) connecting nuclear reactions and β-decays.

of measured Q_m-values exceeds the number of masses contained in them. Auxiliary quantities are not needed here, but the coefficients $\alpha_{\rho l}$ often differ from ± 1.

(*c*) Finally each track in Figure 1 connecting two mass-spectroscopically determined nuclides yields a condition 22 (or a linear combination of them), if one combines the Q_r-, Q_β- and Q_m-values which occur according to equations 4, 7, and 9, taking thereby proper regard of the auxiliary quantities of Table II.

Now, it has been known (3, 8), for some time, that the Q_m-values of the same doublet and also cycles set up according to (b) above often exhibit unexplained discrepancies, even when one is dealing with measurements that the same author made under different conditions. These measurements, therefore, necessarily are distorted by systematic errors. A recent paper by Everling & Mattauch (12) deals with the explanation of such an error source. One knows equally well that conditional relations set up in the way men-

TABLE I

THE PROCEDURE FOLLOWED IN SETTING UP THE SYSTEM OF CYCLES. TESTING THE Q-VALUES FOR ADMITTANCE TO THE LEAST SQUARES ADJUSTMENT WITH THE HELP OF THE VALUES $l_\rho = \epsilon_\rho \left| \sqrt{b_{\rho\rho}} \right|$. ERROR q_I IN kev OF CORRESPONDING Q_I VALUE IS NOTED UNDER EACH REACTION. SEE TEXT

number of cycle old	number of cycle new	cycle		$E_\varrho \pm \sqrt{b_{\varrho\varrho}}$ Mev	l_ϱ	
a	(53)	$^{12}_{3}C(d,n)^{13}N\text{-}^{13}_{3}C(p,n)^{13}N\text{-}^{12}_{3}C(d,p)^{13}C$		± 0,000 ± 0,005	± 0,00	
b	(54)	$^{13}_{80}C(p,\gamma)^{14}N\text{-}^{14}_{1}C(\beta^-)^{14}N\text{-}^{13}_{4}C(d,p)^{14}C$	$+(^1_1n\text{-}^1_1H)\text{-}E_{2^d}$	+ 0,078 ± 0,080	+ 0,98	
c		$^{13}_{160}C(\alpha,n)^{16}O\text{+}^{16}_{2,5}O(d,\alpha)^{14}N\text{-}^{13}_{80}C(p,\gamma)^{14}N$	$+E_{2^d}$	- 0,058 ± 0,179	- 0,32	
d		$^{14}_{200}N(\gamma,n)^{13}N\text{-}^{12}_{3}C(d,n)^{13}N\text{-}^{14}_{80}N(d,\alpha)^{12}C$	$+(E_{\alpha_9}\text{-}2E_d)$	+ 0,030 ± 0,215	+ 0,14	
e	(65)	$^{14}_{80}N(d,\alpha)^{12}C\text{-}^{15}_{3}N(p,\alpha)^{12}C\text{-}^{14}_{7}N(d,p)^{15}N$		- 0,185 ± 0,080	- 2,30	
f		$^{14}_{200}N(\gamma,n)^{13}N\text{-}^{13}_{3}C(p,n)^{13}N\text{+}^{13}_{80}C(p,\gamma)^{14}N$		- 0,077 ± 0,215	- 0,36	
b+c	(57)	$^{12}_{160}C(\alpha,n)^{16}O\text{+}^{16}_{2,5}O(d,\alpha)^{14}N\text{-}^{14}_{1}C(\beta^-)^{14}N\text{-}^{13}_{4}C(d,p)^{14}C$	$+(^1_1n\text{-}^1_1H)$	+ 0,020 ± 0,160	+ 0,13	
f-b	(59)	$^{14}_{200}N(\gamma,n)^{13}N\text{-}^{13}_{3}C(p,n)^{13}N\text{+}^{13}_{4}C(d,p)^{14}C\text{+}^{14}_{1}C(\beta^-)^{14}N$	$-(^1_1n\text{-}^1_1H)\text{+}E_{2^d}$	- 0,155 ± 0,200	- 0,78	
f-b-d		$^{14}_{80}N(d,\alpha)^{12}C\text{+}^{12}_{3}C(d,n)^{13}N\text{-}^{13}_{3}C(p,n)^{13}N\text{+}^{13}_{4}C(d,p)^{14}C\text{+}^{14}_{1}C(\beta^-)^{14}N$	$-(^1_9n\text{-}^1_1H)\text{+}(E_\alpha\text{-}3E_d)$	- 0,185 ± 0,081	- 2,28	
f-b-d-a		$^{14}_{80}N(d,\alpha)^{12}C\text{+}^{12}_{3}C(d,p)^{13}C\text{+}^{13}_{4}C(d,p)^{14}C\text{+}^{14}_{1}C(\beta^-)^{14}N$	$-(^1_9n\text{-}^1_1H)\text{+}(E_\alpha\text{-}3E_d)$	- 0,185 ± 0,081	- 2,28	
f-b-d-a-e	(V)	$^{14}_{7}N(d,p)^{15}N\text{+}^{15}_{3}N(p,\alpha)^{12}C\text{+}^{12}_{3}C(d,p)^{13}C\text{+}^{13}_{4}C(d,p)^{14}C\text{+}^{14}_{1}C(\beta^-)^{14}N$	$-(^1_9n\text{-}^1_1H)\text{+}(E_\alpha\text{-}3E_d)$			
g	(97)	$^{26}_{300}Mg(p,\gamma)^{27}Al\text{-}^{27}_{7}Mg(\beta^-)^{27}Al\text{-}^{26}_{6}Mg(d,p)^{27}Mg$	$+(^1_1n\text{-}^1_1H)\text{-}E_d$	+ 0,187 ± 0,300	+ 0,52	
h		$^{26}_{300}Mg(p,\gamma)^{27}Al\text{-}^{26}_{50}Mg(d,n)^{27}Al$	$-E_{2^d}$	+ 0,493 ± 0,304	+ 1,62	
g-h	(130)	$^{26}_{50}Mg(d,n)^{27}Al\text{-}^{27}_{7}Mg(\beta^-)^{27}Al\text{-}^{26}_{6}Mg(d,p)^{27}Mg$	$+(^1_1n\text{-}^1_1H)$	- 0,336 ± 0,051	- 6,61	

TABLE II

THE AUXILIARY CYCLES FOR THE NEUTRON-PROTON DIFFERENCE AND FOR THE BINDING ENERGIES OF THE PARTICLES USED AS NUCLEAR PROJECTILES. REACTIONS MARKED WITH CROSS-OVER SYMBOLS HAVE *Q*-VALUES ZERO BY DEFINITION. SEE TEXT

No.	auxiliary quantities	auxiliary cycles	measured values Mev ± kev	adjusted values Mev ± kev	
I	$(^1n-^1H)$ =	$^3T(\beta^-)^3He - ^3T(p,n)^3He$ (0.2; 1)	0,782.1 ± 1.0	0,781 4 ± 0.9	
II	E_d =	$^1H(n,\gamma)^2D - \cancel{^1H(n,\gamma)^2D}$ (2)	2,227 ± 2.	2,226 2 ± 1.3	
III	$(E_\gamma-E_d)$=	$^9Be(p,\alpha)^6Li + ^6Li(p,\alpha)^3He + ^3He(\alpha,\gamma)^4He - ^8Be(\gamma,\alpha)^4He - ^9Be(p,d)^8Be$ (2; 2; 1.3; 1)	5,495 5 ± 3.3	5,493 9 ± 2.0	
IV	$(E_\gamma-E_t)$ =	$^3T(p,n)^3He + \cancel{^3He(n,\gamma)^3T}$ (1)	-0,764 ± 1.	-0,763 4 ± 0.9	
V	$[(^1n-^1H)+(E_\alpha-3E_d)]$ =	$^{14}N(d,p)^{15}N + ^{15}N(p,\alpha)^{12}C + ^{12}C(d,p)^{13}C + ^{13}C(d,p)^{14}C + ^{14}C(\beta^-)^{14}N$ (7; 3; 3; 4; 1)	22,394 ± 9.	22,395 ± 6.	
	E_γ =	$^9Be(p,\alpha)^6Li + ^6Li(p,\alpha)^3He + \cancel{^3He(n,\gamma)^4He} - ^8Be(\gamma,\alpha)^4He - ^9Be(\gamma,n)^8Be$ (2; 2; 1.3; 2)	7,719 5 ± 3.7	7,720 1 ± 2.6	
	E_t =	$^9Be(p,\alpha)^6Li + ^6Li(p,\alpha)^3He - ^3T(p,n)^3He + \cancel{^3T(n,\gamma)^4He} - ^8Be(\gamma,\alpha)^4He - ^9Be(\gamma,n)^8Be$ (2; 2; 1; 1.3; 2)	8,483.5 ± 3.8	8,483 5 ± 2.6	
	(E_t-E_d) =	$(E_\gamma - E_d) - (E_\gamma - E_t)$ (3.3; 1)	6,259 5 ± 3.4	6,257 3 ± 1.9	
	$(E_\alpha-3E_d)$ =	$[(^1n-^1H)+(E_\alpha-3E_d)] - (^1n-^1H)$ (9; 1.0)	21,612 ± 9.	21,614 ± 6.	
	$(E_\alpha-2E_d)$ =	$[(^1n-^1H)+(E_\alpha-3E_d)] - (^1n-^1H) + E_d$ (9; 1.0; 2)	23,839 ± 9.	23,840 ± 6.	
	E_α =	$[(^1n-^1H)+(E_\alpha-3E_d)] - (^1n-^1H) + 3E_d$ (9; 1.0; 6)	28,293 ± 11.	28,292 ± 6.	

TABLE III - LEAST SQUARES ADJUSTMENT

		Input Data		Adjusted Values				
I	Process	Q (Mev)	q (kev)	Q* (Mev)	q* (kev)	$v/\|q\|$	l	p
1	2	3	4	5	6	7	8	9
1	$^{1}n(\beta^-)^{1}H$	0.782	13.	0.781 4	0.9	0.04	- 0.01	1
2	$^{2}D(\gamma,n)^{1}H$	-2.227	2.	-2.226 2	1.3	-0.41		
3	$^{2}D(n,\gamma)^{3}T$	6.251	8.	6.257 3	2.0	-0.79	-0.98	2
4	$^{2}D(p,\gamma)^{3}He$	5.50	30.	5.493 9	2.0	0.20	0.15	3
5	$^{2}D(d,p)^{3}T$	4.031	6.	4.031 1	2.0	-0.02	- 0.21	4
6	$^{2}D(d,n)^{3}He$	3.265	9.	3.267 7	2.0	-0.30	- 0.36	5
7	$^{3}T(p,n)^{3}He$	-0.764	1.	- 0.763 4	0.9	-0.62		
8	$^{3}T(p,\gamma)^{4}He$	19.7	200.	19.808 6	4.4	-0.54	-0.53	6
9	$^{3}T(d,\alpha)^{1}n$	17.7	300.	17.582 4	4.3	0.39	0.40	7
10	$^{3}T(\tau,p)^{5}He$	11.15	50.	11.133	29.	0.34	0.25	8
11	$^{3}T(\beta^-)^{3}He$	0.0181	0.2	0.018 06	0.20	0.18		
12	$^{3}He(d,p)^{4}He$	18.45	170.	18.345 8	4.3	0.61	0.63	9
13	$^{3}He(d,\gamma)^{5}Li$	16.3	200.	16.58	70.	-1.38	-1.71	10
14	$^{3}He(\tau,p)^{5}Li$	10.86	150.	11.08	70.	-1.48	-1.82	11
15	$^{5}He(\gamma,n)^{4}He$	0.95	40.	0.956	29.	-0.14		
16	$^{6}He(\beta^-)^{6}Li$	3.50	50.	3.532	26.	-0.64	-0.86	12
17	$^{5}Li(\gamma,p)^{4}He$	1.6	500.	1.77	70.	-0.34	-0.12	13
18	$^{6}Li(\gamma,n)^{5}Li$	-5.37	140.	-5.47	70.	0.70	-0.06	14
19	$^{6}Li(n,\alpha)^{3}T$	4.797	22.	4.786 0	1.9	0.50	0.45	15
20	$^{6}Li(n,d)^{5}He$	-2.57	100.	-2.427	29.	-1.43	-1.37	16
21	$^{6}Li(p,\alpha)^{3}He$	4.023	2.	4.022 6	1.7	0.20		
22	$^{6}Li(d,\alpha)^{4}He$	22.386	11.	22.368 4	4.2	1.60	0.86	17
23	$^{6}Li(d,tp)^{4}He$	2.51	40.	2.559 8	2.0	-1.25	-1.25	18
24	$^{6}Li(d,t)^{5}Li$	0.9	100.	0.79	70.	1.10		
25	$^{6}Li(d,p)^{7}Li$	5.027	3.	5.026 0	1.9	0.32		
26	$^{6}Li(d,n)^{7}Be$	3.40	50.	3.380 8	1.9	0.38	0.37	19
27	$^{6}Li(t,d)^{7}Li$	0.982	7.	0.994 9	2.6	-1.84	-1.46	20
28	$^{6}Li(t,p)^{8}Li$	0.784	15.	0.802 8	2.8	-1.25	-1.17	21
29	$^{7}Li(\gamma,p)^{6}He$	-9.6	300.	-10.003	26.	1.34	1.38	22
30	$^{7}Li(p,\alpha)^{4}He$	17.346	10.	17.342 4	4.3	0.36		
31	$^{7}Li(p,n)^{7}Be$	-1.645 3	0.4	-1,645 26	0.40	-0.10		
32	$^{7}Li(p,\gamma)^{8}Be$	17.1	200.	17.247 4	4.3	-0.74	-0.76	24
33	$^{7}Li(d,\alpha)^{5}He$	14.2	100.	14.161	29.	0.39	0.35	25
34	$^{7}Li(d,p)^{8}Li$	-0.192	1.	-0.192 1	1.0	0.09		
35	$^{7}Li(d,n)^{8}Be$	15,0	100.	15.021 2	4.2	-0.21	0.18	26
36	$^{7}Li(t,\alpha)^{6}He$	9.79	30.	9.806	26.	-0.52		
37	$^{8}Be(\gamma,\alpha\alpha)^{4}He$	0.094 5	1.3	0.095 0	1.2	-0.37		
38	$^{9}Be(\gamma,p)^{8}Li$	-16.93	150.	-16.879 9	4.4	-0.33	-0.30	27
39	$^{9}Be(\gamma,n)^{8}Be$	-1.665	2.	-1,666 6	1.4	0.78	1.00	28
40	$^{9}Be(n,\gamma)^{10}Be$	6.816	6.	6.812 6	3.3	0.57	0.50	29
41	$^{9}Be(p,\alpha)^{6}Li$	2.126	2.	2.125 9	1.6	0.04		
42	$^{9}Be(p,d)^{8}Be$	0.559	1.	0.559 6	0.9	-0.62		
43	$^{9}Be(p,n)^{9}B$	-1.852	2.	-1.852 0	2.0	0.00		
44	$^{9}Be(p,\gamma)^{10}B$	6.5	100.	6.585 9	2.4	-0.86	-0.87	30
45	$^{9}Be(d,\alpha)^{7}Li$	7.153	3.	7.152 0	1.9	0.35		
46	$^{9}Be(d,t)^{8}Be$	4.598	12.	4.590 7	1.9	0.60	0.52	32
47	$^{9}Be(d,p)^{10}Be$	4.588	6.	4.586 4	3.2	0.27	0.39	33
48	$^{9}Be(d,n)^{10}B$	4.35	20.	4.359 7	2.2	-0.48	-0.48	34
49	$^{9}Be(\alpha,p)^{12}B$	-6.92	50.	-6.879	6.	-0.82	-0.61	35
50	$^{10}Be(\beta^-)^{10}B$	0.557	4.	0.554 8	3.1	0.56		

TABLE III - LEAST SQUARES ADJUSTMENT (cont'd.)

		Input Data		Adjusted Values				
I	Process	Q (Mev)	q (kev)	Q° (Mev)	q° (kev)	$v/\|q\|$	l	ρ
1	2	3	4	5	6	7	8	9
51	^{10}B $(\gamma,n)^{9}B$	-8.55	250.	-8.437 9	3.1	-0.45	-0.45	36
52	^{10}B $(n,\alpha)^{7}Li$	2.786	8.	2.792 3	1.7	-0.78	-0.88	37
53	^{10}B $(p,\alpha)^{7}Be$	1.148	2.	1.147 0	1.7	0.50		
54	^{10}B $(p,\gamma)^{8}Be$	-0.536	3.	-0.532 3	2.1	-1.22		
55	^{10}B $(p,n)^{10}C$	-4.35	200.	-4.54	90.	0.93	1.04	39
56	^{10}B $(d,\alpha)^{8}Be$	17.86	40.	17.813 5	4.2	1.16	1.18	40
57	^{10}B $(d,p)^{11}B$	9.235	11.	9.238	5.	-0.30	-2.62	41
58	^{10}B $(d,n)^{11}C$	6.6	100.	6.474	6.	1.26	0.85	42
59	^{10}B $(\alpha,d)^{12}C$	1.39	10.	1.355	6.	3.49		
60	^{10}B $(\alpha,p)^{13}C$	4.08	100.	4.076	6.	0.04	-0.32	44
61	^{11}B $(\gamma,n)^{10}B$	-11.50	250.	-11.465	5.	-0.14	0.01	45
62	^{11}B $(p,\alpha)^{8}Be$	8.585	6.	8.575 1	4.1	1.64		
63	^{11}B $(p,n)^{11}C$	-2.762	3.	-2.764 7	2.8	0.82		
64	^{11}B $(d,\alpha)^{9}Be$	8.018	7.	8.015 5	4.1	0.35	-0.86	46
65	^{11}B $(d,p)^{12}B$	1.137	4.	1.136 5	4.0	0.13		
66	^{11}B $(d,n)^{12}C$	13.8	200.	13.730	5.	0.35	0.37	47
67	^{11}B $(\alpha,p)^{14}C$	0.75	10.	0.778 4	4.5	-2.84	-2.44	48
68	^{12}B $(\beta^-)^{12}C$	13.43	60.	13.375 3	6.4	0.91	0.97	49
69	^{10}C $(\beta^+)^{10}B$	3.8	100.	3.75	90.	0.46		
70	^{11}C $(\beta^+)^{11}B$	1.987	7.	1.963 0	2.9	2.00	2.23	50
71	^{12}C $(\gamma,n)^{11}C$	-19.0	400.	-18.721	6.	-0.70	-0.71	51
72	^{12}C $(n,\gamma)^{13}C$	4.949	6.	4.947 4	2.1	0.27	0.00	52
73	^{12}C $(d,p)^{13}C$	2.722	3.	2.721 2	1.9	0.26		
74	^{12}C $(d,n)^{13}N$	-0.281	3.	-0.281 8	2.3	0.26		
75	^{13}C $(p,n)^{13}N$	-3.003	3.	-3.003 0	2.3	0.00		
76	^{13}C $(p,\gamma)^{14}N$	7.62	60.	7.540 5	3.8	0.99	0.98	54
77	^{13}C $(d,\alpha)^{11}B$	5.164	4.	5.162 0	3.6	0.49		
78	^{13}C $(d,t)^{12}C$	1.310	3.	1.309 9	2.1	0.03		
79	^{13}C $(d,p)^{14}C$	5.942	4.	5.940 4	3.4	0.39		
80	^{13}C $(d,n)^{14}N$	5.17	50.	5.314 3	3.5	-2.89	-2.89	56
81	^{13}C $(\alpha,n)^{16}O$	2.22	160.	2.199 2	4.3	0.13	0.13	57
82	^{14}C $(\beta^-)^{14}N$	0.155	1.	0.155 3	1.0	-0.32		
83	^{13}N $(\beta^+)^{13}C$	2.23	10.	2.221 6	2.4	0.84	0.87	58
84	^{14}N $(\gamma,n)^{13}N$	-10.7	200.	-10.543 5	4.3	-0.78	-0.78	59
85	^{14}N $(n,\alpha)^{11}B$	-0.27	60.	-0.152 3	4.7	-1.96	-1.98	60
86	^{14}N $(n,p)^{14}C$	0.624	4.	0.626 1	1.3	-0.53	-0.73	61
87	^{14}N $(n,\gamma)^{15}N$	10.832	8.	10.842 1	4.2	-1.26	-0.83	62
88	^{14}N $(p,n)^{14}O$	-6.0	200.	-5.934	39.	-0.33	-0.33	63
89	^{14}N $(p,\gamma)^{15}O$	7.34	130.	7.355	6.	-0.12	-0.11	64
90	^{14}N $(d,\alpha)^{12}C$	13.39	80.	13.578 0	4.3	-2.35	-2.30	65
91	^{14}N $(d,p)^{15}N$	8.614	7.	8.615 9	4.1	-0.27		
92	^{14}N $(d,n)^{15}O$	5.12	40.	5.129	6.	-0.23	-0.17	66
93	^{14}N $(\alpha,p)^{17}O$	-1.16	40.	-1.195 5	4.2	0.89	0.89	67
94	^{15}N $(p,\alpha)^{12}C$	4.961	3.	4.962 2	2.7	-0.39		
95	^{15}N $(d,\alpha)^{13}C$	7.681	9.	7.683 4	3.1	-0.26	-0.20	68
96	^{15}N $(d,p)^{16}N$	0.23	150.	0.43	130.	-1.31		
97	^{15}N $(d,n)^{16}O$	10.9	500.	9.832 6	4.8	2.03	2.03	69
98	^{16}N $(\beta^-)^{16}O$	10.5	500.	10.24	130.	0.53	0.13	70
99	^{14}O $(\beta^+)^{14}N$	5.15	40.	5.153	39.	-0.07		
100	^{15}O $(\beta^+)^{15}N$	2.705	5.	2.705	5.	-0.06		

TABLE III - LEAST SQUARES ADJUSTMENT (cont'd.)

		Input Data		Adjusted Values				
I	Process	Q (Mev)	q (kev)	Q° (Mev)	q° (kev)	v/|q|	l	p
1	2	3	4	5	6	7	8	9
101	$^{16}O(\gamma,n)^{15}O$	-15.8	200.	-15.596	7.	-1.02	-1.01	71
102	$^{16}O(d,\alpha)^{14}N$	3.115	2.5	3.115 1	2.5	-0.03		
103	$^{16}O(d,p)^{17}O$	1.919	4.	1.919 6	3.4	-0.15		
104	$^{16}O(d,n)^{17}F$	-1.630	4.	-1.630 9	3.5	0.22		
105	$^{18}O(p,\alpha)^{15}N$	3.961	9.	3.957	8.	0.40		
106	$^{18}O(p,n)^{18}F$	-2.453	4.	-2.452 3	3.9	-0.17		
107	$^{18}O(d,n)^{19}F$	5.7	100.	5.722	9.	-0.22	-0.27	72
108	$^{19}O(\beta^-)^{19}F$	4.5	300.	4.53	280.	-0.08		
109	$^{17}F(\beta^+)^{17}O$	2.771	6.	2.769 0	4.1	0.33	0.49	73
110	$^{19}F(n,\alpha)^{16}N$	-0.73	250.	-1.34	130.	2.43	2.76	74
111	$^{19}F(n,p)^{19}O$	-3.9	750.	-3.74	280.	-0.21	-0.23	75
112	$^{19}F(n,\gamma)^{20}F$	6.63	30.	6.602	7.	0.92	0.97	76
113	$^{19}F(p,\alpha)^{16}O$	8.117	9.	8.118	6.	-0.12		
114	$^{19}F(p,n)^{19}Ne$	-4.039	5.	-4.037 7	4.9	-0.26		
115	$^{19}F(d,\alpha)^{17}O$	10.039	10.	10.038	7.	0.14	0.21	77
116	$^{19}F(d,p)^{20}F$	4.373	7.	4.376	7.	-0.47		
117	$^{19}F(d,n)^{20}Ne$	10.80	200.	10.652	9.	0.74	0.74	78
118	$^{20}F(\beta^-)^{20}Ne$	7.04	20.	7.057	10.	-0.84	-0.81	79
119	$^{19}Ne(\beta^+)^{19}F$	3.21	30.	3.256	5.	-1.54	-1.54	80
120	$^{20}Ne(n,\alpha)^{17}O$	-0.75	50.	-0.614	10.	-2.72	-2.77	81
121	$^{20}Ne(d,\alpha)^{18}F$	2.791	9.	2.788	8.	0.38		
122	$^{20}Ne(d,p)^{21}Ne$	4.528	6.	4.528	5.	0.01		
123	$^{21}Ne(d,\alpha)^{19}F$	6.432	10.	6.434	8.	-0.20		
124	$^{21}Ne(d,p)^{22}Ne$	8.137	11.	8.139	11.	-0.16		
125	$^{22}Ne(d,\alpha)^{20}F$	2.62	100.	2.672	15.	-0.52	-0.47	83
126	$^{22}Ne(d,p)^{23}Ne$	2.964	7.	2.964	7.	0.00		
127	$^{22}Na(\beta^+)^{22}Ne$	2.840	5.	2.840	5.	0.05		
128	$^{23}Na(\gamma,n)^{22}Na$	-12.05	200.	-12.417	14.	1.83	1.83	84
129	$^{23}Na(n,p)^{23}Ne$	-3.6	800.	-3.605	15.	0.01	0.01	85
130	$^{23}Na(p,\alpha)^{20}Ne$	2.378	3.	2.377 8	2.9	0.08		
131	$^{23}Na(p,n)^{23}Mg$	-4.88	10.	-4.875	10.	-0.53		
132	$^{23}Na(d,\alpha)^{21}Ne$	6.902	10.	6.906	5.	-0.37	-0.33	86
133	$^{23}Na(d,p)^{24}Na$	4.727	5.	4.730 6	4.8	-0.71		
134	$^{24}Na(\beta^-)^{24}Mg$	5.511	5.	5.514 5	4.8	-0.71		
135	$^{25}Na(\beta^-)^{25}Mg$	3.7	300.	3.70	290.	0.01		
136	$^{23}Mg(\beta^+)^{23}Na$	3.97	50.	4.093	10.	-2.46	-2.51	87
137	$^{24}Mg(\gamma,n)^{23}Mg$	-16.4	200.	-16.565	12.	0.82	0.81	88
138	$^{24}Mg(n,\gamma)^{25}Mg$	7.334	7.	7.332 3	4.2	0.25		
139	$^{24}Mg(p,\gamma)^{25}Al$	2.14	100.	2.26	50.	-1.21	-1.35	90
140	$^{24}Mg(d,p)^{25}Mg$	5.097	7.	5.106 1	4.2	-1.30		
141	$^{24}Mg(d,n)^{25}Al$	0.07	60.	0.03	50.	0.59		
142	$^{25}Mg(\gamma,p)^{24}Na$	-11.5	1,000.	12.065	6.	0.57	0.56	91
143	$^{25}Mg(d,\alpha)^{23}Na$	7.019	13.	7.044	7.	-1.91	-2.13	92
144	$^{25}Mg(d,p)^{26}Mg$	8.880	10.	8.888	7.	-0.78		
145	$^{26}Mg(\gamma,p)^{25}Na$	-14.0	1,000.	-14.03	290.	0.03	0.02	94
146	$^{26}Mg(\gamma,n)^{25}Mg$	-11.15	200.	-11.114	7.	-0.18	-0.22	95
147	$^{26}Mg(n,\gamma)^{27}Mg$	6.440	8.	6.437 4	4.6	0.32	0.59	96
148	$^{26}Mg(p,\gamma)^{27}Al$	8.4	300.	8.251	7.	0.50	0.52	97
149	$^{26}Mg(d,p)^{27}Mg$	4.207	6.	4.211 3	4.5	-0.71		
150	$^{27}Mg(\beta^-)^{27}Al$	2.591	7.	2.595	6.	-0.55		

TABLE III - LEAST SQUARES ADJUSTMENT (cont'd.)

		Input Data		Adjusted Values				
I	Process	Q (Mev)	q (kev)	Q* (Mev)	q* (kev)	$v/\|q\|$	l	ρ
1	2	3	4	5	6	7	8	9
151	$^{25}Al(\beta^+)^{25}Mg$	4.2	200.	4.29	50.	-0.45	-0.22	98
152	$^{27}Al(n,\gamma)^{28}Al$	7.724	6.	7,723.3	4.8	0.11		
153	$^{27}Al(p,\alpha)^{24}Mg$	1.595	2.	1,595.0	2.0	-0.01		
154	$^{27}Al(p,n)^{27}Si$	-5.610	10.	-5.607	10.	-0.33		
155	$^{27}Al(p,\gamma)^{28}Si$	11.60	90.	11.591	8.	0.10	0.09	99
156	$^{27}Al(d,\alpha)^{25}Mg$	6.694	10.	6.701 1	4.4	-0.71	0.16	100
157	$^{27}Al(d,p)^{28}Al$	5.494	10.	5.497 1	4.9	-0.31	-0.25	101
158	$^{27}Al(d,n)^{28}Si$	9.08	200.	9.364	8.	-1.42	-1.42	102
159	$^{27}Al(\alpha,p)^{30}Si$	2.27	50.	2.380	8.	-2.19	-2.08	103
160	$^{27}Al(\alpha,n)^{30}P$	-2.9	200.	-2.716	30.	-0.92	-0.86	104
161	$^{28}Al(\beta^-)^{28}Si$	4.65	10.	4.649	7.	0.12		
162	$^{27}Si(\beta^+)^{27}Al$	4.70	70.	4.825	10.	-1.79	-1.81	105
163	$^{28}Si(\gamma,n)^{27}Si$	-16.9	200.	-17.197	13.	1.49	1.51	106
164	$^{28}Si(n,\gamma)^{29}Si$	8.468	8.	8.472	5.	-0.54		
165	$^{28}Si(p,n)^{28}P$	-14.9	400.	-14.54	280.	-0.90		
166	$^{28}Si(d,p)^{29}Si$	6.246	10.	6.246	5.	-0.01	0.39	108
167	$^{29}Si(n,\gamma)^{30}Si$	10.601	11.	10.609	6.	-0.71		
168	$^{29}Si(d,\alpha)^{27}Al$	5.994	11.	6.003	7.	-0.82		
169	$^{29}Si(d,p)^{30}Si$	8.388	13.	8.383	6.	0.42	0.82	111
170	$^{29}Si(d,n)^{30}P$	3.27	40.	3.287	30.	-0.42		
171	$^{30}Si(d,\alpha)^{28}Al$	3.120	10.	3.118	7.	0.25		
172	$^{30}Si(d,p)^{31}Si$	4.364	7.	4.369	6.	-0.77		
173	$^{31}Si(\beta^-)^{31}P$	1.473	7.	1.481	6.	-0.87		
174	$^{28}P(\beta^+)^{28}Si$	13.4	400.	13.76	280.	-0.90		
175	$^{30}P(\beta^+)^{30}Si$	4.29	50.	4.314	30.	-0.49	-0.49	112
176	$^{31}P(\gamma,n)^{30}P$	-12.27	120.	-12.391	30.	1.01	1.11	113
177	$^{31}P(n,p)^{31}Si$	-0.94	130.	-0.700	6.	-1.85	-1.90	114
178	$^{31}P(n,\gamma)^{32}P$	7.94	30.	7.934	9.	0.18	0.29	115
179	$^{31}P(p,\alpha)^{28}Si$	1.909	10.	1.916	7.	-0.68		
180	$^{31}P(d,\alpha)^{29}Si$	8.158	11.	8.162	7.	-0.35		
181	$^{31}P(d,p)^{32}P$	5.704	9.	5.708	9.	-0.48		
182	$^{31}P(d,n)^{32}S$	6.81	80.	6.636	9.	2.18	2.23	118
183	$^{32}P(\beta^-)^{32}S$	1.708	4.	1.708 8	4.0	-0.20		
184	$^{32}S(\gamma,d)^{30}P$	-19.15	200.	-19.027	32.	-0.62	-0.53	119
185	$^{32}S(n,\alpha)^{29}Si$	1.16	150.	1.526	12.	-2.44	-2.44	120
186	$^{32}S(n,p)^{32}P$	-0.93	100.	-0.927 4	4.1	-0.03	-0.04	121
187	$^{32}S(n,\gamma)^{33}S$	8.64	20.	8.646	10.	-0.31	-0.39	122
188	$^{32}S(d,p)^{33}S$	6.422	11.	6.420	10.	0.17		
189	$^{32}S(\alpha,p)^{35}Cl$	-2.04	100.	-2.00	70.	-0.42		
190	$^{33}S(d,p)^{34}S$	8.67	250.	8.64	160.	0.11	0.15	123
191	$^{34}S(\gamma,n)^{33}S$	-10.85	200.	-10.67	160.	0.09		
192	$^{35}S(\beta^-)^{35}Cl$	0.167 0	0.4	0.167 01	0.40	-0.02		
193	$^{35}Cl(n,\alpha)^{32}P$	1.02	110.	1.07	70.	-0.46	-0.63	124
194	$^{35}Cl(n,p)^{35}S$	0.52	40.	0.614 4	1.0	-2.36	-2.38	125
195	$^{35}Cl(n,\gamma)^{36}Cl$	8.57	30.	8.563	29.	0.18		
196	$^{35}Cl(d,p)^{36}Cl$	6.28	100.	6.339	29.	-0.59	0.60	126
not admitted:								
197	$^{22}Ne(\alpha,n)^{25}Mg$	-0.916	70.	-0.475	14.	-6.30	-5.93	127
198	$^{23}Ne(\beta^-)^{23}Na$	4.21	20.	4.387	15.	-8.83	-6.69	128
199	$^{23}Na(n,\alpha)^{20}P$	-5.4	300.	-3.898	10.	-5.01	-4.99	129
200	$^{25}Mg(d,n)^{26}Al$	5.58	100.					

TABLE III - LEAST SQUARES ADJUSTMENT (concluded)

		Input Data		Adjusted Values				
I	Process	Q (Mev)	q (kev)	Q* (Mev)	q* (kev)	v/|q|	l	p
1	2	3	4	5	6	7	8	9
201	$^{26}Mg(d,n)^{27}Al$	5.68	50.	6.025	7.	-6.89	-6.61	130
202	$^{27}Al(\gamma,n)^{26}Al$	-14.0	100.					
	"	-12.75	200.					131
203	$^{28}Si(d,n)^{29}P$	0.29	40.					132
204	$^{30}Si(d,n)^{31}P$	4.92	40.	5.069	7.	-3.73	-3.64	133
205	$^{29}P\ (\beta^+)^{29}Si$	4.97	10.					
omitted by mistake :								
a	$^{12}C\ (p,n)^{12}N$	-18.5	100.					
b	$^{12}N\ (\beta^+)^{12}C$	17.6	200.					
c	$^{18}F\ (\beta^+)^{18}O$	1.667	8.	1.670 9	4.0	-0.49	-0.43	

tioned above (*c*) often lead to an inadmissibly large value of *l*. We therefore restrict our least squares adjustment to Q_r- and Q_β-values and discuss in the last section the discrepancy between the adjusted values thus obtained and the mass spectroscopic measurements.

For the sifting of the experimental data all such reactions and β-decays were taken over from the previously mentioned reports (1, 2), put into Table III and drawn in Figure 1, the *Q*-values of which enter at least one cycle of the kind described above (*a*). That means that each mass appears in at least two *Q*-values or that in each point of Figure 1 at least two arrows are beginning or ending. Only those *Q*-values have been omitted which were measured before 1940, or for which only an upper or lower limit is given, or for which the transition to the ground state is not certain and, of course, those which are stated without error, since they have no weight in a least squares adjustment. Inverse reactions, i.e., such reactions which are discerned only by the sign of the *Q*-value are not entered separately; instead, for one of the two reactions the weighted mean together with the error computed in the report by Van Patter & Whaling (1) was taken over. Also in those cases where the same *Q*-value has been measured by more than one author (or group of authors), the weighted mean, together with the error listed in the columns "Average *Q*-value" and "Adopted *Q*" as reported by Van Patter & Whaling (1), and King (2), were taken over. All these *Q*-values and their errors are compiled in columns 3 and 4 of Table III. Since both reports evidently have been prepared with expert knowledge, we did not feel competent to correct in any way the values of the reports, say, on account of different energy standards having been used, etc. Only nine of these *Q*-values numbered $I = 197$ to 205 in Table III have been deemed unfit to be used as input data of the least squares adjustment, for reasons given in the next section.

When setting up the system of cycles (equation 22), one has to take into consideration to begin with, that under polygons also the "diangles" have to be understood, i.e., such two points of Figure 1 which are connected by more than one arrow, e.g., ^{12}C and ^{13}C appearing as target or as end product

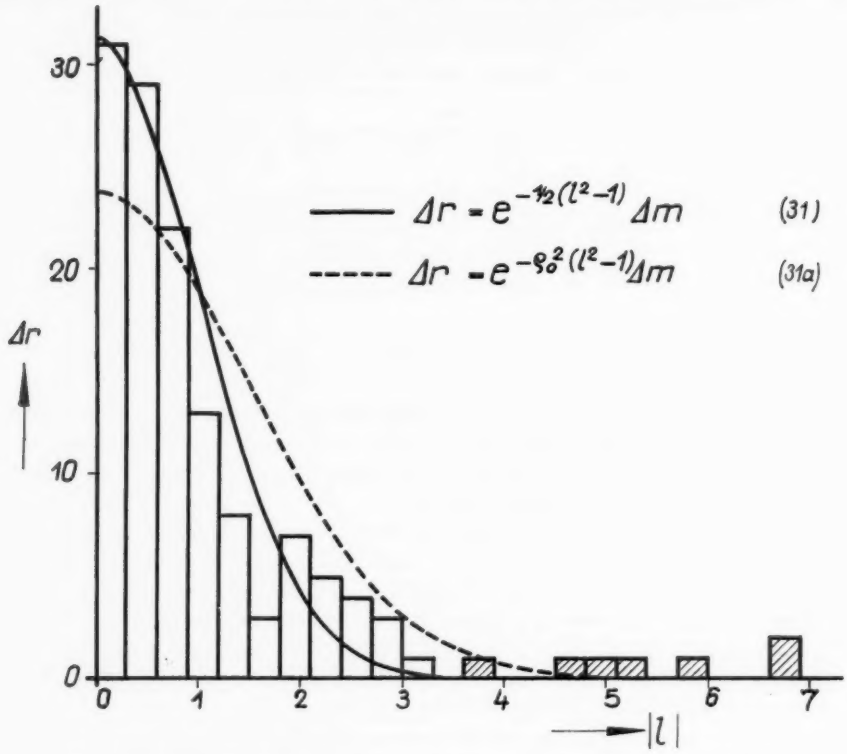


FIG. 2. Distribution of the r cycle-values $|l|$. Δr = number of cycles with l-values between $|l|$ and $|l|+0.3$. (represented by the heights of the rectangles), Δm = number of cycles with l-values between 0.85 and 1.15.

in three reactions of different Q-value yield two cycles, namely $\{^{12}C(n, \gamma)^{13}C - ^{12}C(d, p)^{13}C - E_d\}$ as well as $\{^{13}C(d, t)^{12}C + ^{12}C(d, p)^{13}C - (E_t - E_d) + E_d\}$, which after the least squares adjustment have to be zero. In the same way one may proceed with the triangles, quadrangles, and higher polygons of Figure 1. But in this way one easily obtains much more than r cycles, i.e., they are not linearly independent of each other and above all one is anything but free of arbitrariness in the choice of the r linearly independent cycles.

To do away with this difficulty we demand that the cycles be set up in such a way that each cycle has the smallest possible value of b_{pp}. In other words in the finally accepted system (equation 22), it should not be possible by simple addition and subtraction of cycles to obtain new cycles with smaller values of b_{pp}. Table I serves to illustrate this point. To begin with, there are six cycles listed which are labelled a to f, which may be obtained immediately from Figure 1 by going around the triangles $^{12}C - ^{13}N - ^{13}C$, $^{13}C - ^{14}N - ^{14}C$, $^{13}C - ^{16}O - ^{14}N$, $^{14}N - ^{13}N - ^{12}C$, $^{14}N - ^{12}C - ^{15}N$ and

$^{14}N-{}^{13}N-{}^{13}C$. In order to facilitate the following, the error q_I of the corresponding Q_I-value is noted in kev under each process. Now one tries to eliminate by continuous additive and subtractive combination of these cycles (as shown in column 1 of Table I) such reactions or groups of reactions, which can be replaced by groups of reactions yielding a smaller sum of the squares of errors. For example $(b+c)$ and $(f-b)$ respectively yield the two new cycles $\rho=57$ and $\rho=59$, respectively, which take the place of c and f, respectively. It is obvious that in the new cycles the Q-values of $^{12}C(\alpha, n)^{16}O$ and of $^{14}N(\gamma, n)^{13}N$ are "scrutinized" much more closely, i.e., they are combined with groups of reactions yielding much smaller sums of the squares of errors, than in the old cycles c and d, respectively, where among the "scrutinizing" reactions there are $^{13}C(p, \gamma)^{14}N$ and $^{14}N(d, \alpha)^{12}C$, which have Q-values burdened with the relatively large errors of 80 kev each. The latter reactions had already been "scrutinized" most closely by the cycles b and e, respectively, so that these cycles may be accepted into the final system with $\rho=54$ and $\rho=65$.

The consequent accomplishment of this procedure yields automatically $r+5$ cycles linearly independent of each other. Surplus cycles of the first setting up, i.e., cycles which were not linearly independent of the others, reduce themselves to cycles which are identical with one of these $r+5$ cycles. It is necessary that one obtains just $r+5$ cycles since one needs five equations to evaluate the five auxiliary quantities E_d, E_t, E_τ, E_α and $(^1n-{}^1H)$. For this purpose one chooses those five cycles as auxiliary cycles which yield the five linearly independent combinations of the auxiliary quantities with the smallest values of $b_{\rho\rho}$. They are compiled in Table II and from them other combinations of the auxiliary quantities have been computed as they occur in the now final system (equation 22) of the r conditions. When dealing with polygons in the neighborhood of the origin of Figure 1 it is sometimes useful for formal reasons to add those reactions the Q-values for which are zero by definition. Such reactions are marked by a cross-over sign in Table II. In this way it is easy to make sure that one is dealing with a true cycle. The previously discussed sample of cycles in the upper part of Table I has been chosen in such a way that one can show how the consequent accomplishment of our procedure finally leads to one of the five equations [here denoted by (V)], which has been selected (see Table II) for the computation of the combination $[(^1n-{}^1H)+(E_\alpha-3E_d)]$ of the auxiliary quantities.

Now for each of the r cycles the values of ϵ_ρ, $b_{\rho\rho}$ and l_ρ may be computed according to equations 22, 30, and 29. This is done by inserting for the occurring processes and auxiliary quantities the corresponding values of $Q_I \pm q_I$ from Table III and Table II. Quite often a marked increase of the value of l_ρ results from the reduction of the value of $b_{\rho\rho}$ in the new as compared to the old system of cycles. This becomes evident especially in the example of the lower part of Table I, where the old cycle h is replaced by the new cycle numbered $\rho=130$. The lowering of $\sqrt{b_{\rho\rho}}$ to about $\frac{1}{8}$ of its former value brings about an increase of l_ρ from the quite harmless value 1.62 to the value 6.61, inadmissibly large as we shall see. This ought to make clear,

what is meant by a closer "scrutinizing" of the reaction $^{26}Mg(d, n)^{27}Al$, if we mention that both of the other reactions occurring in the last cycle of Table I have already appeared in former cycles with harmless values of l_ρ.

It is useful to write the cycles in such a way that they always start (with positive sign) with that process which has the Q-value with the largest error q_I. In a prevailing number of cases this process occurs only once; for example the following reaction $^{13}C(p, \gamma)^{14}N$ (see Table I) occurs only in the cycle numbered $\rho = 54$. For these processes the number ρ of the cycle is entered in column nine of Table III. In most of these cases this largest q_I^2 outweighs by far the sum of all other squares of errors occurring in this cycle (e.g., for the cycle $\rho = 54$ it is $6400 \gg 22$) and one may then with good approximation replace $\sqrt{b_{\rho\rho}}$ by q_I. At the same time such a Q-value marked in Table III by the cycle number ρ enters the least squares adjustment with a weight which may be neglected in comparison to the weights of the other Q-values occurring in the same cycle; it will, therefore, suffer the largest deviation v_I, which again will make up the main part of the residual ϵ_ρ. In other words, one may expect that the values l_ρ, so easily computed before carrying through the bulky least squares calculation, will correspond according to sign and size to the values $v_I/|q_I|$, computable only after having carried through the least squares adjustment. Indeed, we shall expect that both quantities entered in columns 7 and 8 of Table III with the sign of v_I and ϵ_ρ, respectively, will agree better the larger the value of $|v_I|$ is, which has been enforced by the adjustment. We shall expect that the Q-value of the reaction $^{13}C(p, \gamma)^{14}N$ will have to suffer a deviation $v_I \approx l_\rho |q_I| \approx \epsilon_\rho \approx 80$ kev by which amount the adjusted value Q_I^* of this reaction will be smaller than the observed value Q_I. The system of reaction cycles (equation 22) set up according to the method described in this section therefore represents, something like a first approximation of the least squares adjustment; it has proved its usefulness as a control of the least squares calculation.

5. The Sifting of The Experimental Data

The answer to the question which maximum value of l_ρ ought still to be admitted to the least squares adjustment is given by the probability integral

$$\Phi(x) = \frac{2}{\sqrt{\pi}} \int_0^x e^{-t^2} dt.$$

The system of r cycles set up in the last section may be compared to r independent observations of one and the same quantity, of which we know the true value, namely zero. For example, in geodesy, the true value of the sum of the angles of a polygon is known before any measurement has been made. However, in our comparison each of the r observations has been made by another method, the precision of which is determined by the standard deviation σ_ρ. The values ϵ_ρ correspond to the true errors of each observations. It is perhaps still more perspicuous to visualize a company of r soldiers, who may be marksmen of different quality. We let each soldier aim one shot at a target and we measure the distance ϵ_ρ between the entrance of the bullet and

the bull's-eye of the target. The standard deviation σ_ρ of each soldier we suppose to be known from previous series of shots. If l is given we have to expect according to theory of probabilities, that $r\Phi(l/\sqrt{2})$ soldiers (cycles) will have hit the bull's-eye closer than is given by l-times their standard deviations, i.e., $\epsilon_\rho < l\sigma_\rho$. Between the values l and $l+\Delta l$ will lie the shots of

$$\Delta r = r\Delta\Phi(l/\sqrt{2}) = r\sqrt{\frac{2}{\pi}}e^{-1/2l^2}\Delta l$$

soldiers (cycles). Finally, if we ascertain by subsequent counting out, the number Δm of soldiers (cycles) with shots in the interval $(1-\frac{1}{2}\Delta l) < l \leqq (1+\frac{1}{2}\Delta l)$, namely $\Delta m = r\sqrt{2/\pi}e^{-1/2}\Delta l$ we obtain the distribution of the soldiers (cycles) among the various values of l

$$\Delta r = e^{-(l^2-1)/2}\Delta m. \qquad 31.$$

In place of the standard deviation (r.m.s. error of a single observation) σ we could have used just as well the probable error of a single observation[4] $\rho = \rho_0\sqrt{2}\sigma = 0.6745\sigma$ as a measure of the precision of the method used, or of the marksmanship of the soldiers. In the consideration above we had then to replace everywhere $1/\sqrt{2}$ by ρ_0 and one obtains instead of equation 31

$$\Delta r = e^{-\rho_0^2\,(l^2-1)}\Delta m. \qquad 31a.$$

The value of $\sqrt{b}$ for each of the conditions on the system (equation 22) is to be identified with σ or with ρ, depending upon whether the errors q_I stated in the literature and in Table III are meant to be r.m.s. errors, or probable errors. The number r of the cycles we have set up is statistically large enough to try to see whether we can differentiate between these two cases, and above all, to find out which ought to be the largest admissible l-value. For the purposes of Figure 2 we have choosen $\Delta l = 0.3$, and have ascertained by counting out that $\Delta m = 19$ cycles have l_ρ-values in the interval $0.85 < l_\rho \leqq 1.15$. This was used to calculate the two curves 31 and 31a of Figure 2, which intersect at the point $l = 1$ and $\Delta r = 19$. Then for each of the successive intervals of size $\Delta l = 0.3$, the number of cycles was ascertained which have values of l_ρ lying in this interval. In the histogram of Figure 2 these numbers are as usual represented by the heights of the rectangles. From Figure 2 one may conclude that the distribution of l_ρ-values for the majority of cycles, especially for $l_\rho < 2$, is better represented by equation 31 than by equation 31a, whereas for a smaller number of instances in which $l_\rho \geqq 2$ the case seems to be reversed. That means that the errors q_I stated by the observers and which we used to bestow weights to each observation, are to be interpreted on the average as something which lies between the r.m.s. and the probable error. In order not to falsify the adjustment unnecessarily by the admission of erroneous measurements or by incorrect weights, it was decided that the largest l_ρ-value admitted should be the one in the interval 3 to 3.3, because curve 31 shows for its occurrence a probability just perceptibly different from zero. The 7 cycles having l_ρ-values larger than

[4] The (transcendental) number ρ_0 is, as is well known, defined by $\Phi(\rho_0) = \frac{1}{2}$.

3.3 (the corresponding rectangles in Fig. 2 have been hatched), were erased from the system, and the 9 values $Q_I \pm q_I$ occurring only in the erased cycles and numbered $I = 197$ to 205 in Table III have not been used as input data for the least squares adjustment. With that, the number of values Q_I admitted as input data for the least squares adjustment turns out to be $N = 196$; they comprise $n = 70$ masses and are related by $r = 126$ linearly independent cycles.

For the sake of completeness we have to confess that, by a mistake, 3 Q-values (1, 2) have been omitted, namely those of $^{12}C(p, n)^{12}N$, of $^{12}N(\beta^+)^{12}C$, and of $^{18}F(\beta^+)^{18}O$. The first two processes would have yielded a new mass, namely that of ^{12}N, however, the error would have been relatively large because these two processes are stated with errors of 100 and of 200 kev. respectively. The Q-value of $^{18}F(\beta^+)^{18}O$ is stated with an error of 8 kev. It would have only slightly ameliorated the mass difference $^{18}F - ^{18}O$, which for the main part is determined by the Q-value of the reaction $^{18}O(p, n)^{18}F$ which is stated with an error of 4 kev only.

6. Details of The Least Squares Calculation

One starts by setting up the rectangular scheme (matrix) of the stoichiometric coefficients κ_{Ii}, which has $N = 196$ rows and $n = 70$ columns corresponding to the Q_I-values of the 196 processes and to the masses M_i of the 70 nuclides. The co-ordination of the nuclides has been done by increasing Z and within the same element by increasing A, as is seen from Table IV. The sequence of processes according to target or β-ray emitter and for the same target according to projectiles was the sequence of Table III which follows closely the order used in the reports (1) and (2). As far as the coefficients κ_{Ii} differ from zero, they have nearly always the values $+1$ or -1, which makes their scheme very simple. The first six columns for ^1n, ^1H, ^2D, ^3T, ^3He and ^4He are occupied quite densely by coefficients differing from zero, since the projectiles of the reactions reappear again and again. For the rest only the neighborhood of the diagonal of the rectangular scheme is occupied by coefficients differing from zero, since fission and spallation processes do not occur. (Ed. note: see page 214, Footnote[6] for Errata in Tables IV and V).

As far as given by Li *et al.* (6) their masses were used as approximate values $M_i^{(0)}$, and for the rest they were computed from the Q-values with smallest errors as pointed out in Section 3. The computation of the values $Q_I^{(0)}$ according to equation 12 or that of the λ_i-values according to equation 14 consists then of simple multiplications of one row or of one column of the scheme of coefficients κ_{Ii} with the values of $M_i^{(0)}$ or of $(Q_I^{(0)} - Q_I)q_I^{-2}$, respectively, whereby the summation is done automatically by the storage part of the calculating machine. The computation of the terms of the matrix (a_{ik}) according to equation 14 is almost as simple and can be done easily by a skilled operator. However, for each a_{ik} the weights $1/q_I^2$ have to be multiplied with the κ-values of two different columns, the ith and the kth column, and the products be summed up. The matrix (a_{ik}) has 70 rows and 70 columns and is symmetrical, i.e. $a_{ik} = a_{ki}$. It contains terms differing from

zero only in the first six rows and columns as well as in the neighborhood of the diagonal.

The part of the calculation which is the most complicated by far, and which surpassed the calculating facilities of our Institute is, of course, the computation of the inverse matrix (a^{-1}_{ik}). We are very grateful that it was done for us first at the calculating institute of the Technische Hochschule,

TABLE IV – ADJUSTED MASS EXCESSES & BINDING ENERGIES

1	Nuclide	M*(Mev)	m*(kev)	M*(mMU)	m*(μMU)	E*(Mev)	e*(kev)
1	2	3	4	5	6	7	8
1	^{1}n	8.365 8$_{1}$	1.6	8.984 2$_{7}$	1.7	-	-
2	^{1}H	7.584 3$_{6}$	1.7	8.145 0$_{5}$	1.8	-	-
3	^{2}D	13.724 0	2.7	14.738 6	2.9	2.226 2	1.3
4	^{3}T	15.832 5$_{0}$	4.2	17.002 9$_{5}$	4.5	8.483.4$_{8}$	2.6
5	^{3}He	15.814 4$_{4}$	4.2	16.983 5$_{5}$	4.5	7.720 1$_{0}$	2.6
6	^{4}He	3.608 2	2.0	3.875 0	2.1	28.292	8
7	^{5}He	12.930	29	13.886	31	27.336	29
8	^{6}He	19.393	27	20.827	29	29.239	26
9	^{8}Li	12.96	70	13.92	70	26.52	70
10	^{6}Li	15.860 9	4.4	17.033 5	4.7	31.990	7
11	^{7}Li	16.974 $_{52}$	6	18.229 $_{39}$	6	39.241 $_{81}$	7
12	^{8}Li	23.306 $_{2}$	7	25.029 $_{2}$	7	41.278 $_{9}$	8
13	^{7}Be	17.838 $_{33}$	6	19.157 $_{06}$	6	37.596 $_{55}$	7
14	^{8}Be	7.311 6	3.9	7.852 0	4.2	56.489	10
15	^{9}Be	14.010 7	4.9	15,046	5	56.186	11
16	^{10}Be	15.564 $_{0}$	7	16.714 $_{6}$	7	64.968 $_{3}$	12
17	^{9}B	15.081 $_{3}$	5	16.196 $_{2}$	6	56.303 $_{8}$	11
18	^{10}B	15.009 $_{2}$	5	16.116 $_{8}$	6	64.741 $_{6}$	12
19	^{11}B	11.910 5	4.6	12,791	5	76.206	15
20	^{12}B	16.913 $_{6}$	7	18,164 $_{0}$	7	79.568 $_{8}$	16
21	^{10}C	18.76	90	20.15	100	60.21	90
22	^{11}C	13.893 $_{5}$	6	14.920 $_{6}$	6	73.441 $_{7}$	15
23	^{12}C	8.538 4	3.6	8.800 0	3.9	92.163	19
24	^{13}C	6.956 8	3.8	7.471 1	4.1	97.110	19
25	^{14}C	7.156 0	2.9	7.685 0	3.1	105.277	21
26	^{13}N	9.178 4	4.5	9.856 9	4.8	94.107	19
27	^{14}N	7.000 7	2.8	7.518 2	3.0	104.651	21
28	^{15}N	4,524 4	4.2	4.858.9	4.5	115.493	23
29	^{16}N	10,24	130	10,99	130	118.15	130
30	^{14}O	12.153	39	13.052	42	98,716	45
31	^{15}O	7,230	6	7,764	7	112.006	24
32	^{17}O	4,220 0	3.6	4.532.0	3.9	131.747	26
33	^{18}O	4,508 $_{7}$	9	4,838 $_{8}$	9	139.827 $_{3}$	29
34	^{19}O	8.67	280	9.31	300	144.03	280
35	^{17}F	6.989 1	3.8	7.505 7	4.1	128.197	26
36	^{18}F	6.176 $_{6}$	9	6.633 $_{2}$	10	137.374 $_{9}$	29
37	^{19}F	4.141 $_{9}$	7	4.448 $_{1}$	7	147.775 $_{4}$	31
38	^{20}F	5.905	9	6.342	10	154.378	32
39	^{19}Ne	7.398 $_{2}$	8	7.945 $_{1}$	9	143.737 $_{7}$	31
40	^{20}Ne	-1.151 $_{5}$	9	-1.236 $_{7}$	10	160.653 $_{2}$	34
41	^{21}Ne	0.460	10	0.494	10	167.407	35
42	^{22}Ne	-1.539	15	-1.653	16	171.772	37
43	^{23}Ne	1.637	16	1.758	18	182,962	38
44	^{22}Na	1,301	15	1.397	17	174.151	38
45	^{23}Na	-2,749 $_{9}$	10	-2.953 $_{2}$	11	186.567 $_{6}$	39
46	^{24}Na	-1.340 $_{8}$	11	-1.439 $_{9}$	12	193.524 $_{3}$	40
47	^{25}Na	-2.12	290	-2.28	310	194.31	290
48	^{23}Mg	1.343	14	1.443	15	181.693	40
49	^{24}Mg	-6.855 $_{4}$	12	-7.362 $_{2}$	13	198.257 $_{4}$	41
50	^{25}Mg	-5.821 $_{8}$	13	-6.252 $_{2}$	14	205.589 $_{7}$	42

TABLE IV - ADJUSTED MASS EXCESSES & BINDING ENERGIES (Concluded)

i	Nuclide	M*(Mev)	m*(kev)	M*(mMU)	m*(μMU)	E*(Mev)	e*(kev)
1	2	3	4	5	6	7	8
51	^{26}Mg	-8.570_0	14	-9.203_5	15	216.703_7	44
52	^{27}Mg	-6.641_6	14	-7.132_6	15	223.141_1	45
53	^{25}Al	-1.53	50	-1.65	60	200.52	60
54	^{27}Al	-9.236_5	13	-9.919_3	14	224.954_8	45
55	^{28}Al	-8.594_0	14	-9.229_3	15	232.677_6	46
56	^{27}Si	-4.411	16	-4.737	17	219.348	46
57	^{28}Si	-13.243	15	-14.222	16	236.545	48
58	^{29}Si	-13.349	15	-14.336	16	245.017	49
59	^{30}Si	-15.592	16	-16.745	17	255.63	50
60	^{31}Si	-13.822	16	-14.844	18	262.22	50
61	^{28}P	0.52	280	0.56	300	222.00	290
62	^{30}P	-11.278	33	-12.112	36	250.53	60
63	^{31}P	-15.303	15	-16.434	18	262.92	50
64	^{32}P	-14.671_{17}	19	-15.971_2	20	270.86_6	50
65	^{32}S	-16.890_6	19	-17.806_3	20	271.78_3	50
66	^{33}S	-16.861	21	-18.108	23	280.43	60
67	^{34}S	-19.36	160	-20.79	170	291.30	170
68	^{35}S	$-18.39_{1\cdot37}$	80	$-19.75_{1\ 20}$	80	$298.69_{1\ 75}$	90
69	^{35}Cl	$-18.55_{6\cdot58}$	80	$-19.93_{0\ 56}$	80	$298.07_{7\ 31}$	90
70	^{36}Cl	-18.75_7	80	-20.14_4	90	308.64_2	100

Darmstadt, Germany (Professor Walther) with a Holerith-machine under the direction of Professor Unger, and, after we had made a few small changes in the matrix (a_{ik}), a second time at the Max-Planck-Institut für Physik, Göttingen, Germany with the G2, the larger of their two electronic machines (Professor Biermann), under the direction of Dr. Jörgens. The matrix $(a^{-1}{}_{ik})$ is, of course, also symmetrical. The terms $a^{-1}{}_{ik}$ for $i \neq k$ change in sign just as do the terms a_{ik}, originating from the changing sign of the coefficients κ_{Ii}. Their magnitude varies even in the same row or column by several powers of ten, because the errors q_I, from which they descend in the end, spread over a range of more than three powers of ten. However, the first peculiarity which makes operating with the inverse matrix much more wearisome and complicated than with the matrix (a_{ik}) is the fact that all the terms differ from zero. Already the calculation of a single mass amelioration, which according to equation 15 is

$$\delta M_i^* = M_i^{(0)} - M_i^* = \sum_k a^{-1}{}_{ik}\lambda_k, \qquad 32.$$

requires 70 multiplications and the summation of the products. The second and still more aggravating peculiarity is the high number of digits required for both factors of equation 32. From Göttingen we got the $35 \times 71 = 2485$ terms $a^{-1}{}_{ik}$ differing from each other and with up to ten digits each. At first sight this high number of digits seems very striking since the errors q_I are stated with only one, or at the utmost, two digits; however, it was necessary for the calculating institutes themselves for carrying out their controls which consisted in multiplications of rows and columns. In the computation of the amelioration δM_i^* according to equation 32, often very small terms $a^{-1}{}_{ik}$ were paired with very large λ_k-values and vice versa; furthermore, the products differ in sign, and the summation ended up in the difference of two

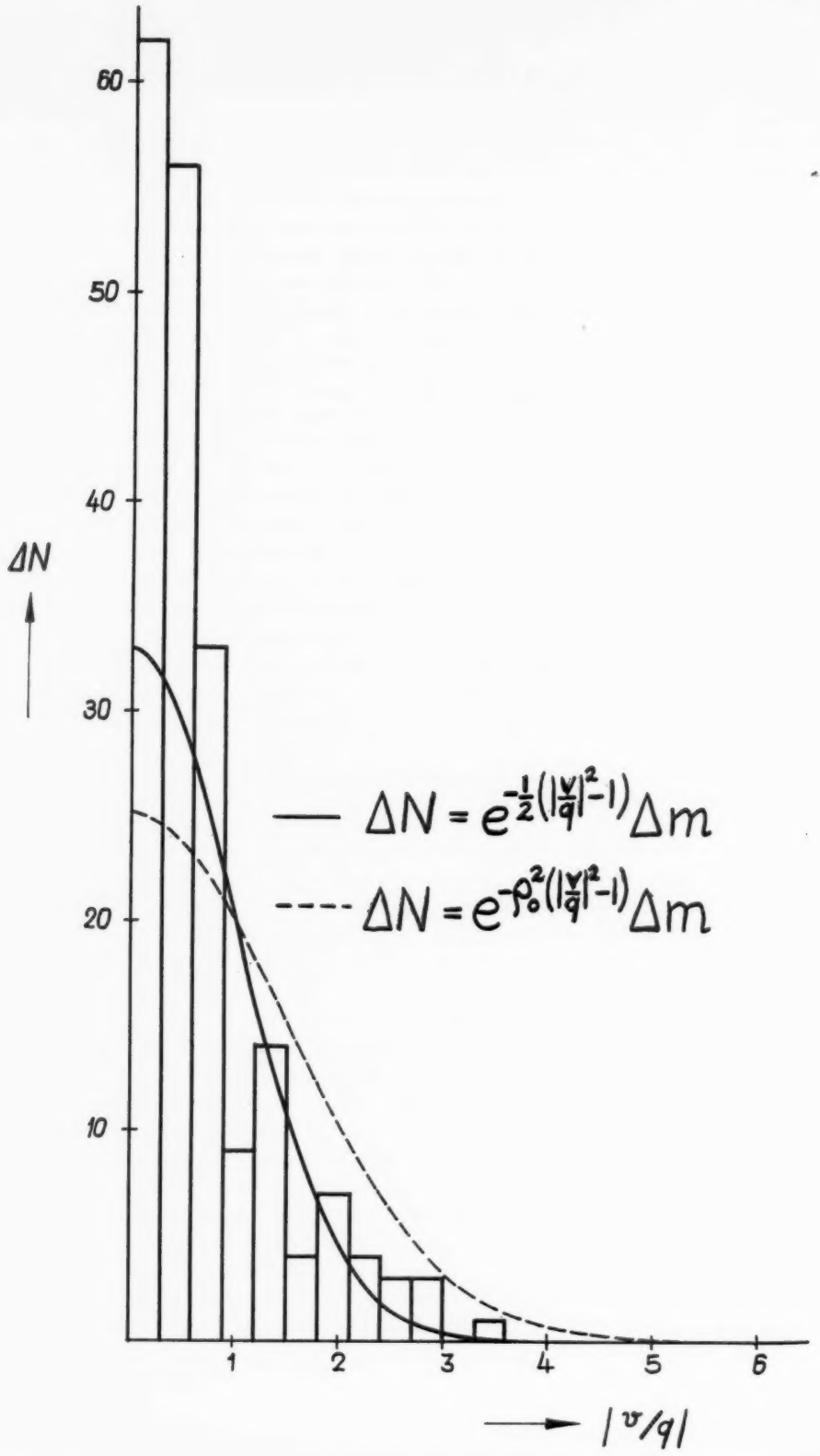
nearly equal large numbers. A closer inspection of the inverse matrix and of the λ-values showed that 4 to 8 digits have to be required of each factor in order that the ameliorations δM_i^* be falsified by less than about 0.01 kev by the rounding off errors. This requirement was made, because the smallest adjusted error, namely that of $^3T(\beta^-)^3He$, is $q^* = \pm 0.20$ kev and errors originating by the calculation should vanish compared with it.

Parallel to the compution of the ameliorations δM_i^*, which, when subtracted from the approximate masses $M_i^{(0)}$ yields at once the adjusted masses M_i^*, the adjusted errors m_i^* (the square roots of the terms of the diagonal) and q_I^* according to equation 17 could be computed, since one needs for it besides the known stoichiometric coefficients, only the knowledge of the terms $a^{-1}{}_{ik}$. To enable users of this report to calculate the adjusted error of any desired linear combination Q^* of the masses M^*, e.g., of mass spectroscopic doublet values, of diverse binding energies, etc., the matrix $(a^{-1}{}_{ik})$ is reproduced in an abridged but for this purpose adequate form in Table V. Taking into account the power of ten at the head of each column, one gets each term in (kev)^2. In each column we trimmed the number of digits according to the diagonal term, mostly the largest term of the column, which was written down with 3 digits. Therefore, terms of the same column vanish if they are smaller by three powers of ten. This accuracy of the terms $a^{-1}{}_{ik}$ suffices for the computation of adjusted errors q^* in most cases, but not always. For, the inverse matrix has the property that corresponding terms of two rows or columns are of nearly equal size, if those rows or columns are co-ordinated to two nuclides which are connected by an especially precisely measured process. This holds particularly for the rows and columns co-ordinated to the pairs $^3T - {}^3He$, $^7Li - {}^7Be$, and $^{35}S - {}^{35}Cl$; because $^3T(\beta^-)^3He$, $^7Li(p, n)^7Be$, and $^{35}S(\beta^-)^{35}Cl$ were the sole processes which have been entered into the least squares adjustment with an error q_I smaller than 1 kev. For example, although the adjusted masses of ^{35}S and of ^{35}Cl each have the relatively high adjusted error $m^* = \pm 76$ kev, their difference was known from the β-decay of ^{35}S with an error of only $q = \pm 0.4$ kev even before the least squares calculation was started. According to equations 17 and 18 the square of the adjusted error of the difference $^{35}S - {}^{35}Cl$ is given by

$$m^{*2}(^{35}S) + m^{*2}(^{35}Cl) - 2a^{-1}(^{35}S, {}^{35}Cl), \qquad 33.$$

i.e., from the sum of the squares of the two errors (which then would yield the square of the error of the mass-difference, provided the two masses had been measured independent of each other), there is to be subtracted the correlation term of practically equal size. Even if we use four digits of each of the three matrix terms occurring in equation 33 we get for the square of the adjusted error of $^{35}S(\beta^-)^{35}Cl$ the value zero, which makes no sense. Only by using six digits one obtains 0.16 (kev)^2 and with it not even a trifling reduction of the adjusted error compared to the error mentioned above of ± 0.4 kev.

Each adjusted error q_I^* in Table III has to be smaller than the corresponding value q_I, if sometimes only by a trifling amount; because each


60
50
40
ΔN
30
20
10
1
2
3
4
5
6
$\Delta N = e^{-\frac{1}{2}(|\frac{v}{q}|^2-1)}\Delta m$
$\Delta N = e^{-\rho_0^2(|\frac{v}{q}|^2-1)}\Delta m$
$|v/q|$

adjusted value Q_I^* is composed of more information than given by the input value Q_I alone. This fact and the comparison mentioned at the end of section 4 between the columns headed v/q and l in Table III gave us the first possibilities to control the computation of the inverse matrix. We received very valuable hints as to the parts of the inverse matrix where one had to search for errors of calculation when we were in possession of the first trial solution of Darmstadt. This had been kindly delivered to us even before the calculating institute had carried out its own control and it was computed to fewer digits than was done later in Göttingen. The most important and extensive control could be taken up only after the ameliorations δM_i^* had been computed. Solving equation 32 we get

$$\lambda_i = \sum_k a_{ik}\delta M_k^*. \qquad 34.$$

Substituting our values of δM^* into equation 34 and comparing the values of λ_i thus obtained with those computed according to equation 14 controls in a very sensitive way the computation of the inverse matrix $(a^{-1}{}_{ik})$ from the matrix (a_{ik}), as well as the calculation of the ameliorations δM_i^* from the terms $a^{-1}{}_{ik}$ and the values λ_i. This test proved that the adjusted masses M_i^* had been calculated to better than about 0.01 kev.

The computation of the ameliorations δM_i^*, of the adjusted errors q_I^*, and the above mentioned test had kept several, at first untrained calculators busy for over two months. The calculation of the quantities given in Tables III and IV, including the binding energies $E_i^* \pm e_i^*$, is then straightforward. We mention that, for the conversion of the masses from Mev into mMU the conversion factor given by DuMond and Cohen (4) 1 mMU = 0.931162 Mev has been used, the error of which may be here neglected.

7. Results of the Least Squares Adjustment and Comparison with Mass-Spectroscopic Measurements

In Table III and IV the adjusted errors q_I^*, m_i^* and e_i^* are rounded off as usual to two digits if the first figure differing from zero is 1 to 4, and to one digit if it is 5 to 9. In Table III the adjusted values Q_I^* have been rounded off to the decimal place corresponding to the last valid digit of the attached error q_I^*. As a rule the values M_i^* and E_i^* of Table IV have been treated in the same way; however, in many cases an extension of 1 to 3 digits (printed in low placed brevier) had to be made here so that the values Q_I^* of Table III could be computed to the decimal place mentioned above. This was done because linear combinations of M_i^*- or of E_i^*-values, especially some of their differences, are quite frequently known with far greater accuracy than the masses or binding energies themselves. This has been shown already in the

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Fig. 3. Distribution of the N values $|v/q|$ of Table III. ΔN = number of data with $|v/q|$-values between $|v/q|$ and $|v/q| + 0.3$ (represented by the heights of the rectangles), $2\Delta m$ = number of data with $|v/q|$-values between 0.7 and 1.3.

preceding section taking as an example the β^--decay of ^{35}S. If according to the adjusted error of 80 kev the masses of ^{35}S and of ^{35}Cl, respectively, had been rounded off to -18.39 Mev and to -18.56 Mev, respectively, then the total β-disintegration energy of ^{35}S could have been computed to two digits only, namely 0.17 Mev. From the statements in Table IV, however, with the help of the 3 low printed digits the adjusted value of this disintegration energy can be stated as 0.16701 Mev corresponding to the adjusted error of 0.40 kev.

TABLE V. THE MATRIX a^{-1}_{ik} OCCURRING IN LEAST SQUARES ADJUSTMENT

Terms are in $(kev)^2$

		10^{-2}	10^{-2}	10^{-2}	10^{-1}	10^{-1}	10^{-2}	10^{0}	10^{0}	10^{1}	10^{-1}	10^{-1}	10^{-1}	10^{-1}	10^{-1}	10^{-1}	10^{-1}	10^{-1}
1n	1	257								448								
1H	2	233	295							2	192							
2D	3	383	425	754						2	225	307						
3T	4	532	589	1035	173					3	272	370	462					
3He	5	530	591	1036	173	174				2	229	309	369	320				
4He	6	228	259	430	58	58	396			1	146	178	211	183	152			
5He	7	623	645	1145	180	180	734	836		2	196	243	294	246	182	237		
6He	8	681	928	1711	276	276	976	30	706	2	242	309	375	313	212	280	432	
6Li	10	549	593	1057	162	163	733	18	28	2	200	245	294	257	188	240	284	292
7Li	11	702	724	1350	203	203	882	23	37	2	225	307	372	316	206	270	346	276
8Li	12	852	885	1680	248	248	1085	28	45	3	272	370	462	390	252	330	422	276
7Be	13	678	786	1391	209	209	912	23	37	2	229	309	369	320	206	270	346	211
8Be	14	422	478	806	114	114	739	14	20	1	146	178	211	183	152	177	260	211
9Be	15	581	612	1122	160	160	904	19	28	2	196	243	294	246	182	237	280	432
10Be	16	727	765	1421	208	208	1068	24	37	2	242	309	375	313	212	280	432	199
9B	17	557	674	1165	166	166	935	20	29	2	200	245	294	257	188	240	284	292
10B	18	676	741	1369	208	208	1027	24	37	2	238	309	372	316	206	270	346	276
11B	19	424	454	831	121	121	600	14	21	1	136	176	213	178	121	159	195	162
12B	20	572	582	1156	165	165	770	19	29	2	184	237	294	238	154	210	260	211
10C	21	671	753	1378	209	209	1033	24	37	2	239	310	372	318	207	270	347	279
11C	22	404	507	866	126	126	627	14	21	1	142	177	213	187	126	162	199	172
12C	23	180	187	231	29	29	218	4	5	0	41	52	59	55	45	53	62	55
13C	24	285	276	493	73	73	364	6	13	1	83	105	126	107	74	95	118	97
14C	25	212	217	431	61	61	211	7	10	1	61	61	102	82	42	63	82	64
13N	26	244	341	561	79	79	406	9	13	1	90	112	134	121	81	102	126	112
14N	27	197	208	408	58	58	188	6	10	1	56	75	95	76	38	57	75	58
15N	28	180	175	341	45	45	352	6	6	1	65	82	98	81	70	86	101	85
16N	29	240	206	471	63	63	371	8	12	1	79	103	129	99	73	98	119	94
14O	30	196	210	409	58	58	189	6	10	1	57	75	95	76	38	57	75	59
15O	31	183	179	348	46	46	352	6	9	1	66	83	99	82	70	86	101	86
17O	32	147	147	345	47	47	186	5	8	0	49	65	85	65	36	54	69	54
18O	33	126	75	238	29	29	378	5	6	0	58	71	86	66	73	87	98	82
19O	34	29	3	81	9	9	209	2	2	0	28	33	40	30	40	46	50	44
17F	35	132	178	364	49	49	197	5	8	1	80	65	83	69	37	54	69	59
		1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17
		1n	1H	2D	3T	3He	4He	5He	6He	5Li	6Li	7Li	8Li	7Be	8Be	9Be	10Be	9B
		10^{-2}	10^{-2}	10^{-2}	10^{-1}	10^{-1}	10^{-2}	10^{0}	10^{0}	10^{1}	10^{-1}	10^{-1}	10^{-1}	10^{-1}	10^{-1}	10^{-1}	10^{-1}	10^{-1}
18F	36	92	125	248	32	32	384	5	6	1	58	68	81	72	74	85	96	89
19F	37	25	12	87	10	10	214	2	2	0	28	33	40	31	41	47	51	46
20F	38	151	122	357	47	47	373	6	9	1	66	86	109	83	71	92	108	89
19Ne	39	2	72	128	16	16	244	2	3	0	32	35	41	42	46	50	55	47
20Ne	40	-111	-104	-190	-29	-29	239	-1	-4	0	8	-4	-13	-3	46	37	28	37
21Ne	41	3	-13	58	5	5	361	3	2	0	39	43	50	41	69	74	77	78
22Ne	42	149	114	380	49	48	528	7	10	1	85	104	131	101	101	124	141	121
23Ne	43	299	244	709	93	93	699	12	17	1	131	167	213	161	134	178	207	170
22Na	44	149	113	379	49	48	528	7	10	1	85	104	131	100	101	124	141	121
23Na	45	-118	-143	-192	-30	-30	374	0	-4	0	18	11	6	8	71	65	57	62
24Na	46	-16	-57	42	1	1	506	3	2	0	52	57	67	52	97	104	106	100
25Na	47	16	-45	80	6	6	573	4	3	0	62	68	81	62	109	119	123	113
23Mg	48	-142	-84	-152	-25	-24	403	0	-3	0	22	13	6	18	76	68	60	74
24Mg	49	-62	-101	-53	-12	-12	471	2	0	0	40	40	48	36	90	92	90	88
25Mg	50	44	-11	126	13	13	597	5	4	1	68	76	90	71	113	126	130	119
26Mg	51	-62	-122	-31	-9	-10	569	3	1	0	52	54	63	48	108	114	113	108
27Mg	52	67	-11	205	23	22	714	6	6	1	87	100	121	92	136	153	153	146
25Al	53	89	110	320	39	39	682	7	8	1	92	105	126	107	130	147	160	149
27Al	54	-89	-129	-35	-10	-11	616	3	1	0	56	58	67	51	117	123	122	116
28Al	55	125	34	284	30	29	795	8	7	1	98	113	135	103	151	169	180	160
27Si	56	-83	-69	5	-5	-5	645	3	1	0	60	60	67	61	122	126	126	127
28Si	57	-100	-177	-144	-26	-26	626	2	-2	0	47	44	47	36	119	119	113	111
29Si	58	-7	-100	7	-5	-5	740	4	2	1	71	75	85	65	140	148	148	136
30Si	59	25	-80	31	-2	-2	805	5	2	1	79	83	95	73	152	160	161	150
31Si	60	77	-42	175	17	17	900	7	6	1	102	114	135	102	170	188	195	176
28P	61	-112	-146	-123	-23	-23	641	2	-2	0	49	45	47	41	121	120	115	117
30P	62	49	-11	185	19	19	862	7	6	1	98	108	128	102	163	178	186	172
31P	63	-20	-133	- 10	-8	-9	825	5	1	1	78	82	94	70	156	164	164	153
32P	64	138	7	324	37	37	1001	10	9	1	125	145	177	131	190	216	230	203
32S	65	138	7	325	37	37	1001	10	9	1	125	145	177	131	190	216	230	203
33S	66	313	161	666	84	83	1155	15	17	2	174	209	260	194	225	269	286	254
34S	67	528	354	1028	134	133	1391	21	26	2	226	276	344	259	263	324	357	307
35S	68	122	-1	348	39	39	1153	11	10	1	141	161	196	149	218	247	262	234
35Cl	69	122	-1	348	39	39	1153	11	10	1	141	161	196	149	218	247	262	234
36Cl	70	370	223	726	92	91	1376	17	19	2	195	231	281	216	260	304	334	289
		1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17
		1n	1H	2D	3T	3He	4He	5He	6He	5Li	6Li	7Li	8Li	7Be	8Be	9Be	10Be	9Be

TABLE V (continued)

		10^{-1}		
3T	4	173,484	10^{-1}	
3He	5	173,447	178,809	
		4	5	
		3T	3He	

| | | | |

TABLE V (concluded)

		10^1															
18F	36	795	10^1														
19F	37	189	428	10^1													
20 F	38	236	422	851	10^1												
19Ne	39	192	427	419	677	10^1											
20Ne	40	524	304	360	305	900	10^1	10^0									
21Ne	41	465	340	393	338	775	906	212	10^0								
22Ne	42	475	348	421	344	762	907	214	268	10^0							
23Ne	43	467	350	448	350	784	914	215	268	237	10^0						
22Na	44	474	348	421	344	762	906	212	214	237	81	10^0					
23Na	45	544	327	366	325	925	823	81	81	81	106	106	10^0				
24Na	46	556	336	406	331	923	823	84	86	84	106	129	106	10^2			
25Na	47	565	341	415	336	932	845	85	87	85	107	127	635	10^0			
23Mg	48	547	326	383	321	926	821	81	80	81	106	105	1	201	10^0		
24Mg	49	556	336	402	331	930	835	84	84	84	106	127	1	106	147	10^0	
25Mg	50	566	341	416	335	931	843	85	87	85	107	126	2	106	144	159	10^0
26Mg	51	574	349	421	343	952	861	87	87	87	110	130	2	109	149	154	199
27Mg	52	584	355	437	347	949	868	88	91	88	110	131	2	109	151	154	186
28Al	53	570	340	421	342	922	839	86	86	86	105	127	1	106	147	146	180
27Al	54	562	355	428	346	964	873	88	89	88	111	133	2	111	153	151	159
28Al	55	592	360	443	351	988	876	89	92	89	111	133	2	110	153	152	159
27Si	56	585	354	428	355	964	871	87	88	87	111	132	2	111	152	151	158
28Si	57	589	361	430	353	982	886	88	89	89	114	138	2	113	155	153	161
29Si	58	598	367	442	357	984	894	90	91	90	114	136	2	114	156	155	163
30Si	59	606	372	448	362	995	905	91	92	91	116	137	2	115	158	157	164
31Si	60	617	380	464	368	998	916	93	95	93	117	139	2	116	159	158	166
28P	61	591	360	428	357	982	885	88	89	88	114	134	2	113	155	153	161
30P	62	610	373	456	367	987	903	92	94	92	115	137	2	114	157	156	164
31P	63	615	380	457	369	1009	919	93	94	93	118	140	2	117	160	159	167
32P	64	627	387	480	374	1000	926	95	98	95	117	140	2	116	160	160	168
32S	65	627	387	480	374	1000	926	95	96	95	117	140	2	116	160	160	168
33S	66	639	393	502	379	991	931	98	103	98	117	141	2	115	161	161	169
34S	67	649	398	520	381	981	934	100	106	100	116	141	2	114	160	162	169
35S	68	655	407	504	395	1035	962	99	103	99	122	146	2	121	166	166	175
35Cl	69	655	407	504	395	1035	962	99	103	99	122	146	2	121	166	166	175
36Cl	70	664	410	520	395	1024	963	101	106	101	121	146	2	120	165	167	174
		36	37	38	39	40	41	42	43	44	45	46	47	48	49	50	51
		18F	19F	20F	19Ne	20Ne	21Ne	22Ne	23Ne	22Na	23Na	24Na	25Na	23Mg	24Mg	25Mg	26Mg

		10^1															
25Al	53	263	10^0														
27Al	54	15	164	10^0													
28Al	55	15	164	189	10^0												
27Si	56	15	163	163	261	10^0											
28Si	57	15	167	183	168	229	10^0	10^0									
29Si	58	16	168	183	167	214	226	214	10^0								
30Si	59	16	170	186	169	209	214	241	267	10^2							
31Si	60	16	172	190	171	216	219	235	267	802	10^1						
28P	61	15	166	183	166	229	213	209	215	802	110	10^0					
30P	62	16	169	186	169	212	221	224	227	2	110	266	10^0				
31P	63	16	173	190	172	223	223	228	247	2	23	266	343	10^0			
32P	64	16	174	192	173	223	224	229	249	2	23	267	343	358	10^0		
32S	65	16	174	192	173	223	224	229	249	2	23	267	343	358	459	10^2	
33S	66	16	174	194	173	223	225	230	251	2	23	268	345	361	459	249	10^1
34S	67	16	174	195	173	222	225	230	253	2	23	268	347	363	462	4	584
35S	68	17	181	199	180	231	232	237	258	2	24	276	352	360	363	4	585
35Cl	69	17	181	199	180	231	232	237	258	2	24	276	352	360	363	4	584
36Cl	70	17	180	200	179	230	232	238	259	2	24	276	364	362	367	4	585
		53	54	55	56	57	58	59	60	61	62	63	64	65	66	67	68
		25Al	27Al	28Al	27Si	28Si	29Si	30Si	31Si	28P	30P	31P	32P	32S	33S	34S	35S

(Fig. 2). All the statements of error (before and after adjustment) of Table III and IV could now be multiplied by the factor 1.18 or 0.80, respectively, in order to get intervals which are adequate to what is understood by the r.m.s. or the probable error, respectively. We have refrained from doing this, so that the adjusted errors stated in our tables may just correspond to that interval which, on the average is apparently felt by the various observers to be "the error."

In analogy to section 5 and the histogram of Figure 2 one can also investigate the distribution of the N values $|v_I/q_I|$ replacing everywhere $|l|$ by $|v/q|$ and r by N. However, contrary to what has been said before no normal (Gaussian) distribution is to be expected now, because the N input data Q, or their adjusted values Q^*, respectively, are not linearly independent of each other as were the r cycles; therefore the comparison made in section 5 does not hold here and the distribution cannot be determined by the probability integral. Indeed, the histogram of Figure 3 shows a very marked deviation from the curves (computed with $\Delta m = 20$) no matter whether the q_I are taken to be the r.m.s. or the probable errors. It is interesting to observe that according to Gauss's condition, equation 10, the least squares adjustment works in such a way that in an overwhelming majority the values $|v/q|$ are pressed into the intervals next to the origin.

A survey of the variation of the adjusted error with the mass number A is given by the hatched regions of Figures 4 and 5 which indicate the interval of error of the packing fraction M^*/A and of the binding energy per nucleon E^*/A. Since with isobaric nuclides the errors frequently differ quite appreciably, the nuclide with the smallest error m^* or e^*, respectively, was chosen at each mass number for the purposes of the two figures. This is nearly always the stable one which is quite clear since it is more easily accessible to measurement than the unstable isobars. Exceptions are only the mass numbers $A = 5$ and $A = 8$ for which there are no stable nuclides and for which ^{5}He and ^{8}Be have been chosen, as well as $A = 36$ which in our system of 70 masses is represented by the unstable ^{36}Cl only.

To begin with Figure 5 we notice that the error e^*/A of the binding energy per nucleon, being zero by definition at $A = 1$, very rapidly approaches a constant value (± 1.6 kev). The boundary of the hatched region shows little chiseling with the exception of two striking peaks due to ^{5}He and to ^{34}S as well as a broadening caused by ^{35}Cl and ^{36}Cl. These peculiarities can easily be understood. According to equation 2, $E_i^*/A_i \approx \frac{1}{2}(^1n^* + {}^1\mathrm{H}^*) - M_i^*/A_i$ neglecting thereby entirely the term $T_{\zeta i}(^1n^* - {}^1\mathrm{H}^*)/A_i$ as we may for the following computation of errors. Analogous to equation 3 we split the square of the error of E^*/A computed according to equation 17 into the two squares of error and the correlation term by writing $(e_i^*/A_i)^2 = q^{*2} + (m_i^*/A_i)^2 - K_i$. Here the abbreviations $q^{*2} = \frac{1}{4}(a^{-1}{}_{11} + a^{-1}{}_{22} + 2a^{-1}{}_{12})$ and $K_i = (a^{-1}{}_{1i} + a^{-1}{}_{2i})/A_i$ are introduced. Compared to $q^{*2} = 2.54$ (kev)^2 the two other terms may be neglected as a rule. $q^* = \pm 1.6$ kev determines the constant value of e^*/A mentioned above and represents the error of $\frac{1}{2}(^1n^* + {}^1\mathrm{H}^*)$, i.e., according to

equation 5 the error of the binding energy per nucleon at the standard ^{16}O. Only if the mass of the nuclide is very inaccurately known so that $m^{*2} = a^{-1}_{ii}$ is very large, then the second term appreciably enlarges the generally constant value of $(e^*/A)^2$. Perusing Figure 1 and Table III one learns that ^5He and ^{34}S and to a lesser degree also ^{35}Cl and ^{36}Cl are connected with nuclides of neighboring mass number by inaccurately known reactions only. That is, their Q-values are stated with errors q_I which are large compared to the errors of Q-values which have served for the computation of the masses of these neighboring nuclides. As an example ^{34}S is connected with ^{33}S by two reactions only, the Q-values of which are stated as 200 and 250 kev, whereas errors of 11 and 20 kev are attached to the Q-values of the two reactions leading from ^{32}S to ^{33}S. Indeed, the four nuclides mentioned which gave rise to the striking peaks and to the broadening of the otherwise constant interval of error exhibit values of a^{-1}_{ii} which are larger by two to four powers of ten than the terms of the first two columns of the inverse matrix. The correlation term K_i is markedly noticeable only at very small values of A; it diminishes, as a rule, the square of the error. With it the error is gradually reduced for nuclides lighter than ^{16}O until for ^2D the value $e^*/A = \pm 0.6$ kev is reached.

The hatched region of Figure 4 indicating the variation of the error m^*/A of the packing fraction with mass number A presents in many respects a somewhat different picture. We disregard here entirely the above mentioned four nuclides having exceptionally large values of m^*; they have the same effect here as shown in Figure 5. In general the error of the packing fraction is considerably smaller than the error of the binding energy per nucleon since the error of $\frac{1}{2}(^1n^* + {}^1H^*)$ governing Figure 5 does not appear. Starting with ^{16}O where the error of the packing fraction is zero by definition m^*/A rises about proportionally with decreasing A, reaching at ^1H the value ± 1.7 kev which practically coincides with the value of the error q^* of Figure 5. However, the carving or chiseling of the boundary of the hatched region is here much more pronounced. The tight lacings at ^2D, ^4He and ^8Be are especially striking. The masses of the first two nuclides are very well known by numerous accurately measured (d, α) reactions. ^8Be is the end product of several reactions measured with high precision and above all it is connected with ^4He by its very well known decay into two α-particles. For nuclides with $A > 16$ until about ^{33}S, m^*/A approaches quickly a practically constant value, i.e., the gradual increase of the error of measuring Q-values in this region is about compensated by the factor $1/A$.

For the comparison of Table VI between the adjusted masses computed from nuclear data (Q_r- and Q_β-values) and those from mass spectroscopic doublet measurements (Q_m-values) only those of the latter have been taken into consideration which have been made since the publication of the Duckworth, *et al.* paper (3). Because of the discrepancies mentioned and known for some time, it was to be expected that in the mean time further efforts have been made to avoid as far as possible systematic errors with mass

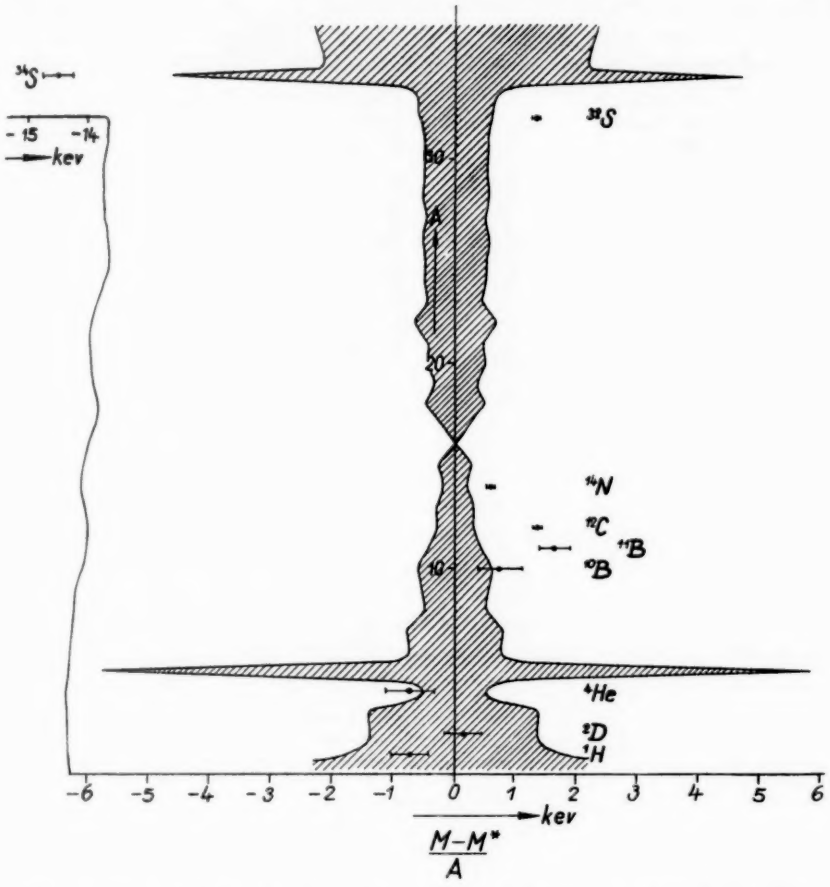

FIG. 4. The hatched region shows the variation of the error $\pm m^*/A$ of the packing fraction M^*/A with mass number A.

The differences $(M-M^*)/A$ of mass spectroscopically determined masses M as compared to the adjusted masses M^* from nuclear physics data are entered as small circles. The limited tracts indicate the errors of mass spectroscopic measurement, i.e. m/A.

spectroscopic measurements. The measurements by Scolman, Quisenberry & Nier (13), by Smith (14) and by Schierstedt, Ewald, Liebl & Sauermann (15) are collected in the pairs of columns (5, 6), (7, 8) and (9, 10), respectively, of Table VI denoted by $Q \pm q$ and numbered $I=1$ to 15. For comparison the values $Q^* \pm q^*$ of these mass differences computed from the adjusted masses of Table IV have been entered into the pair of columns (3, 4). The second part of Table VI contains accordingly the mass spectro-

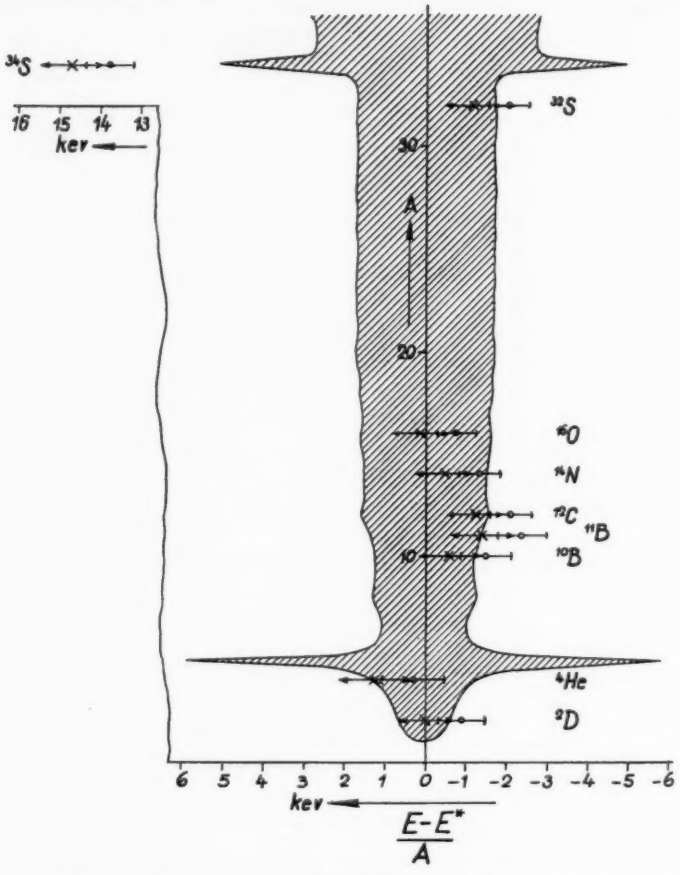

FIG. 5. The hatched region shows the variation of the error $\pm e^*/A$ of the binding energy per nucleon E^*/A with mass number A.

The differences $(E-E^*)/A$ of binding energies E calculated from mass spectroscopic measurements with the help of two different assumptions concerning the mass of the neutron as compared to the adjusted values E^* from nuclear physics data are entered as small circles or as crosses, respectively.

scopic mass excesses $M \pm m$ calculable from the measured doublets of each paper and again for comparison the adjusted values $M^* \pm m^*$ from Table IV.

Some remarks have to be made concerning the data in Table VI. Wapstra[5] has made a complete least squares solution of the set of doublet measurements given in the abstracts published by Nier *et al.* (13) by which the masses concerned are heavily overdetermined. He calls attention to the fact that

[5] A. H. Wapstra, private communication. We are very grateful to Dr. Wapstra for his permission to make use here of the results of his least squares adjustment.

this adjustment results in $R_e/R_i = 4.1$ instead of the expectation value 1 ± 0.2 and is, therefore, somewhat invalidated by the presence of systematic errors or by the fact that too high a precision of measurement has been claimed. Nevertheless, Wapstra's adjusted values were entered into Table VI because otherwise an unambiguous comparison would not have been possible. Wapstra's adjusted values differ from those given by Nier *et al.* (13) in most cases by a larger error since, as is usual in such a case, these adjusted errors have been multiplied by R_e/R_i. The M-values differ by amounts which are always smaller than this error. The mass difference Q_{12} was not measured directly by Nier *et al.*, but is easily computed from the adjusted masses. To facilitate comparison the mass differences Q_6, Q_{11}, Q_{12} and Q_{14} in the column headed Smith were computed from the other directly measured doublet values. Three of the latter had been adjusted with the help of the cycle $2Q_3 - Q_4 + Q_5 = 0$ whereby the measured values stated by Smith (14) were altered by fractions of the error only. Ewald *et al.* (15) have measured two to three series at different times of each of the doublets Q_4, Q_7, Q_8 and Q_9. The weighted means of these which are listed in Table VI differ from the means given by Ewald *et al.* (15) practically only by a somewhat smaller error. The remaining three mass differences of this column have been computed from these doublets for the sake of comparison.

A comparison of the values listed in the columns headed Nier *et al.* and Smith shows quite astonishing agreement far within the very small error statements at all Q- and M-values measured by both authors. This agreement is the more valuable since the measurements have been made, as is well known, by two entirely different methods. In Minneapolis the new large double focusing mass spectrometer developed by A. O. Nier has been used and in Brookhaven L. G. Smith takes advantage of his modern and original mass synchrometer. These mass spectroscopic measurements differ, however, in many particulars from the results of our least squares adjustment of nuclear physics data. In order to work out clearly the agreement or the discrepancy, respectively, of the mass spectroscopic values M with the masses M^* of our least squares adjustment the differences $(M - M^*)/A$ by which the mass spectroscopic packing fractions M/A differ from the values M^*/A have been entered as small circles into Figure 4. The limited tracts indicate the errors of the mass spectroscopic measurement, i.e., m/A. Weighted means of the results of both authors have been used for ^{1}H, ^{2}D, ^{12}C, ^{14}N and ^{32}S, whereas Smith alone is responsible for ^{4}He, ^{10}B and ^{11}B. There is agreement at the three lightest nuclides of which ^{1}H and ^{4}He show negative differences. All the heavier nuclides exhibit positive differences and from ^{11}B on the discrepancy with the hatched region is markedly pronounced. Since $T_\nu = 0$ for most nuclides measured mass spectroscopically up to now, one can by no means consider annuling the discrepancy by altering the adjusted values M^* through the introduction of a neutrino mass differing from zero. Also an error of the conversion factor cannot be blamed since the discrepancies $(Q - Q^*)$ of the positive doublet values do not always have the same sign.

TABLE VI - Comparison of New Mass-Spectroscopic Doublet Measurements ($Q \pm q$) and of Masses ($M \pm m$) Derived Thereof with the Corresponding Values ($Q^* \pm q^*$, $M^* \pm m^*$) of the Least Squares Adjustment of Nuclear Physics Data

I	Doublet	Adjusted values		Nier, et al (1)[a]		Smith (14)		Ewald, et al	
		Q*(mMU)	q*(μMU)	Q(mMU)	q(μMU)	Q(mMU)	q(μMU)	Q(mMU)	q(μMU)
1	2	3	4	5	6	7	8	9	10
1	$(^1H_2-^2D)$	1.551 5	1.6	1.549 8	0.4	1.549 3	0.4		
2	$(^2D_2-^4He)$	25,602 1	4.4			25.609 0	0.5		
3	$(^{12}C^1H_2-^{14}N)$	12.572	5	12.575 9	0.6	12.579 1[b]	0.6		
4	$(^{12}C^1H_4-^{16}O)$	36.380	9	36.394 1	1.3	36.394 8[b]	1.5	36.371 4[c]	4.4
5	$(^{14}N_2-^{12}C^{16}O)$	11.236	5	11.236 3	1.4	11.236 7[b]	1.5		
6	$(^{14}N^1H_2-^{16}O)$	23.608	6	23.613 2	1.1	23.615 7	1.3		
7	$(^{16}O_2-^{32}S)$	17.806	20	17.761 3	3.2	17.761 6	2.4	17.754 [c]	6
8	$(^{32}S^1H_2-^{34}S)$	19.26	170					19.551 [c]	8
9	$(^{12}C_3-^{34}S^1H_2)$	15.90	170					16.456 [c]	7
10	$(^{12}C_4-^{32}S^{16}O)$	33.006	24	33.030 5	3.1	33,0308	7.2	32.968	11
11	$1/2(^{12}C_4{}^{16}O-^{32}S_2)$	25.406	21	25.395 9	2.6	25.396 2	3.8	25.370	6
12	$(^{16}O_2{}^1H_4-^{12}C_3)$	21.180	12	21.125	5	21.126	8	21.140	12
13	$1/5(^{11}B_5{}^1H_9-^{32}S^{16}O_2)$	21.013	5			21.022 4	2.6		
14	$(^{11}B^1H-^{12}C)$	17.136	6			17.137 3	3.2		
15	$(^{10}B^1H-^{11}B)$	11.473	6			11.460 8	1.9		
i	Mass Excess	M*(mMU)	m*(μMU)	M(mMU)	m(μMU)	M(mMU)	m(μMU)	M(mMU)	m(μMU)
I	$(^1H-1/16^{16}O)$	8,145 0	1.8	8,144 2	0.4	8,144 4	0.6	8.1409	1.1
II	$(^2D-1/8^{16}O)$	14,738 6	2.9	14.738 6	0.8	14,739 6	1.3		
III	$(^4He-1/4^{16}O)$	3,675 0	2.1			3,670 3	2.6		
IV	$(^{10}B-5/8^{16}O)$	16,118	6			16,126 7	3.9		
V	$(^{11}B-11/16^{16}O)$	12,791	5			12,810 2	3.1		
VI	$(^{12}C-3/4^{16}O)$	3,800 0	3.9	3,817 3	1.2	3,817 3	1.9	3.807 9	3.2
VII	$(^{14}N-7/8^{16}O)$	7,518 2	3.0	7.526 8	1.0	7,527 0	1.2		
VIII	$(^{32}S-2\ ^{16}O)$	-17.806	20	-17.761 3	3.2	-17.761 6	2.4	-17.754	6
IX	$(^{34}S-17/8^{16}O)$	-20.79	170					-21.324	9

a) All values in this column according to least squares solution made by A. H. Wapstra.
b) Adjusted with the help of the cycle $2Q_3-Q_4+Q_5=0$.
c) Weighted mean values of the series given in the paper.
d) Entry for $I=$ is not a measured doublet, but is computed from Q-values of the same column.

From the column headed Ewald *et al.* one gets in kev the values -3.9 ± 2.0, $+0.57\pm0.38$ and $+1.5\pm0.6$ for the differences $(M-M^*)/A$ of the nuclides ^1H, ^{12}C and ^{32}S which are to be compared with the values (see points in Figure 4) -0.74 ± 0.32, $+1.35\pm0.08$ and 1.30 ± 0.06, resulting from the weighted means of the masses listed in the columns headed Nier *et al.* and Smith. It seems, therefore, to us that Ewald *et al.* (15) got the agreement of their ^{12}C mass with nuclear physics data (a point repeatedly stressed by Ewald) at the price of an exceptionally low mass of H^1, whereas at ^{32}S the difference is rather the same or larger. The new measurements of the

Munich group were made by taking great care to avoid systematic errors. Nevertheless, they seem to show a general trend compared to the values from nuclear physics data which runs in the same direction as do the measurements of the American authors stated with much smaller errors. In principle this is brought out by the mass difference Q_{12} which has been computed by Ewald *et al.* (15) themselves and which, contrary to Q_4, contains the masses of ^{1}H and of ^{12}C not in the same term. All mass spectroscopic measurements including those of Ewald *et al.* exhibit here practically the same discrepancy to the least squares adjustment of nuclear physics data. However, Ewald *et al.* (15) proceed in their paper in a different way. With the help of a value $^1\text{H} = (8.144 \pm 0.001)$ mMU, which is arrived at as the mean of the measurements of other authors and which for this reason agrees with the values of the American authors listed in Table VI, the mass of ^{12}C is computed from Q_{12} with the result $^{12}\text{C} = (3.812 \pm 0.006)$ mMU. But with these masses one gets $Q_4 = (36.388 \pm 0.007)$ mMU in quite good agreement with the values of the American authors. On the other hand, Ewald *et al.* (15) emphasize again the good agreement of their new low value of Q_4 with former measurements of their own so that it is not clear why they did not use it to calculate the mass of ^{1}H.

For ^{34}S only the new measurement of Ewald *et al.* is listed in Table VI. The difference $(M - M^*)/A$ entered in Figure 4 has the abnormally great value -14.49 ± 0.24 kev. As is seen from Figure 1 ^{34}S lies at the end of a small chain and is connected with ^{33}S only by the reactions $^{33}\text{S}(d, p)^{34}\text{S}$ and $^{34}\text{S}(\gamma, n)^{33}\text{S}$. Our adjusted mass ^{34}S* rests on the Q-values of these two reactions. They are stated with errors of 250 and of 200 kev and they appear (cf. Table III) in the cycle $\rho = 123$ with $l_\rho = 0.15$, in other words, the sum of the two Q-values agrees far within the interval of error with the value of $-E_d$ from Table II. The mass spectrographic measurement of Ewald *et al.* requires that the absolute values of the Q's of both reactions should be raised by 500 kev; that means that by the investigation of the reaction $^{33}\text{S}(d, p)$ not the ground level of ^{34}S was reached but an excited level lying higher by about 500 kev and, furthermore, that accidentally the threshold of the reaction $^{34}\text{S}(\gamma, n)$ has been measured too low by about the same amount. Wapstra (8) has shown in detail that all mass spectroscopic and microwave measurements made up to now as well as a few nuclear physics data stated without error are in favor of this interpretation. Fortunately, the deletion of ^{34}S* from our system of masses can hardly influence the matrix (a^{-1}_{ik}) and the results of our least squares adjustment. Because ^{34}S is only connected with ^{33}S by two Q-values of negligibly small weights and no other mass is derived from it.

When comparing packing fractions, the discrepancies $(M - M^*)/A$ necessarily vanish at the standard ^{16}O lying in the middle of Figure 4. However, if one wishes to compute binding energies from mass spectroscopic data he has to make some assumption about the mass of the neutron. The following two seem to offer themselves. (*a*) We accept the neutron-proton difference of the least squares adjustment $(^1n^* - {}^1\text{H}^*) = 0.7815$ Mev ± 0.9 kev (in the

following the magnitudes are stated in Mev and the errors in kev), i.e., we put the main weight on the Q_β-values. With it and with the weighted means of the masses of ^{1}H and of ^{2}D from the data of Nier *et al.* and of Smith we get $(^1n+^1\mathrm{H}) = 15.9487 \pm 1\,1$, $^1n = 8.3651 \pm 1.0$ and $E_d = 2.2244 \pm 1.2$. (*b*) We let the adjusted binding energy of the deuteron $E_d^* = 2.2262 \pm 1.3$ unaltered which may mean that we give the preference to the Q_r-values. Then we get $(^1n - ^1\mathrm{H}) = 0.7832 \pm 1.3$, $(^1n + ^1\mathrm{H}) = 15.9505 \pm 1.4$ (practically coinciding with the adjusted value $(^1n^* + ^1\mathrm{H}^*) = 15.9502 \pm 3.2$) and $^1n = 8.3668 \pm 1.3$. With these values and with the mass spectroscopic masses from Table VI the binding energies per nucleon E/A have been computed according to equation 1 or 2 and their differences $(E - E^*)/A$ to the values of the least squares adjustment have been entered into Figure 5.

From assumption (*a*) there follow the differences marked as little circles with the limited tracts attached characterizing the errors and from assumption (*b*) the ones marked as crosses with the attached tracts limited by arrow points. The discrepancies seem to be not at all as pronounced in Figure 5 as they were in Figure 4. However, this somewhat deceiving agreement was obtained at the price of assumptions which would require more or less small alterations of all nuclear physics data and, therefore, also of the values E^*.

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[6] Errata: (*a*) Table 4: line 1-47, column 7 should read 202.64 instead of 194.31. (*b*) Table 5, the headings of columns 35 to 41 should read 10^{-1} instead of 10^1.

NUCLEAR RADIATION EFFECTS IN SOLIDS

By H. Brooks

Division of Engineering and Applied Physics, Harvard University, Cambridge, Massachusetts

INTRODUCTION

The last article in this series dealing with radiation damage was prepared by Dienes (1) and covered the literature up to February 1, 1952. During the intervening four years several other reviews have appeared (2, 3, 4, 5, 18). In August of 1955, the International Conference on the Peaceful Uses of Atomic Energy was held under U. N. auspices at Geneva.[1] At this conference, there were two sessions on radiation damage; one dealing primarily with the basic science, and the other with materials of direct practical interest for reactors. The papers prepared for these two sessions by authors from the U. S., Britain, the U.S.S.R., and France together constitute a rather comprehensive summary of the whole subject. The session dealing with reactor materials was of special interest because in it information on uranium and graphite, accumulated in several countries since the beginning of the atomic program, was publicly discussed for the first time. Although much of this information is not new, it has not been previously available in the open literature, and it therefore seems appropriate to devote a portion of this review to it.

In general the period since Dienes' review has been characterized by the growth of a body of quantitative and detailed knowledge of the radiation and annealing behavior of a few simple materials, most notably copper and germanium. The extension of such studies to irradiations at liquid nitrogen and more recently at near liquid helium temperatures has led to a realization of the extremely high mobility of the lattice defects produced. For example, annealing effects are observed below 30°K., and the shape of the damage curve as a function of integrated particle flux depends on temperature even in the vicinity of 10°K. (6).

In spite of the wealth of new experimental information, however, the damage to simple metals and alloys is not well understood, and in particular there is no general agreement as to the identity of the imperfections responsible for the various annealing processes. Indeed, the identification of the lattice imperfections responsible for property changes remains the central problem of the theory of radiation damage. For the solution of this problem it is fruitful to carry out comparative studies of the effects of heat treatment, cold work, and irradiation damage, and this program has been followed in the case of Cu, Ge, and a number of simple binary alloys. The use of alterna-

The survey of literature pertaining to this review was completed in February, 1956.

[1] Papers cited from this conference are listed in the references as Geneva Paper 5 (1955).

tive techniques for introducing imperfections has also proved useful in studies of graphite and of alkali halides.

The literature of radiation damage has grown by leaps and bounds since the Dienes (1) review. It has proved impossible to cover every aspect of the subject within the confines of a short review. Such an inclusive review would reduce to little more than a catalog. Accordingly only representative topics from each of the major fields of investigation have been selected for somewhat more detailed discussion.

The article begins with a review of the status of the theory of atomic displacements, with special emphasis on recent modifications and on critical experimental tests of the theory. In this section, I have leaned heavily on recent reviews, especially that of Seitz & Koehler (9), to whom I am indebted for an advance copy of their manuscript. The second section of the article is devoted to the recently declassified results on graphite and uranium. Here I have relied heavily on the Geneva papers, since most of the original literature is in the form of previously classified reports which are not yet generally available. The third section is devoted to detailed reviews of recent work on metals and alloys, valence semiconductors, other valence crystals, and ionic crystals. Relatively little attention has been paid to experimental techniques, although in many cases they are of critical importance. The general types of techniques in the field have been well covered in Dienes' article.

THEORY OF ATOMIC DISPLACEMENTS

The theory of radiation damage naturally divides itself into three parts (7). These are (*a*) the production of displaced atoms and other lattice imperfections by bombarding particles, (*b*) the nature and mobility of the imperfections and their role in annealing and recovery, and (*c*) the relation between radiation-produced imperfections and observable properties. The present section will be concerned mainly with item (*a*).

General Theory

The main features of the theory of the production of displaced atoms have been understood for some years (8). Recently a comprehensive and critical review of this topic has been made by Seitz & Koehler (9). The modifications of the original theory are mostly in matters of quantitative detail. The main new developments are (*a*) the realization that throughout the energy ranges encountered in practice, a classical rather than a quantum mechanical description of the collision process is more appropriate (10, 22); (*b*) a more accurate estimate of the collision radius in the "hard sphere" range of collisions between knocked-on atoms (9), and (*c*) an improved theory of the number of secondary displacements produced by a primary knocked-on atom (3, 9, 22). The remaining uncertainties in the theory stem from the estimate of the "hard sphere" collision radius, and from the lack of an adequate theory of energy loss by electronic excitation and ionization for very slow charged particles.

The interaction between atoms ~~or between atoms~~ or between charged particles and atoms may be described in terms of a screened Coulomb field. The screening parameter ζ determines whether the scattering is governed by the exponentially tailing part of the field or by the Coulomb part. For collisions between particles having nuclear charges Z_1 and Z_2 and masses M_1 and M_2 it is given by:

$$\zeta = Z_1 Z_2 (Z_1^{2/3} + Z_2^{2/3})^{1/2} \left(1 + \frac{M_1}{M_2}\right) \frac{2R_h}{E}$$

where R_h is the ionization energy of hydrogen.

Weak screening always applies in the case of bombardment by charged particles from accelerators, while strong screening is applicable to the collisions of knocked-on atoms with the lattice for $Z > 10$. Classical mechanics is always valid for knocked-on atoms, and usually for accelerator particles. Collisions with electrons usually require a wave-mechanical description, and for the cases of practical interest the electrons are in the relativistic range.

In order that all the collisions of the knocked-on atoms be of the hard sphere type it is necessary that strong screening apply over their entire range. For the maximum energy transferred to an atom by a 2 Mev neutron equation (1) requires that $Z > 26$, assuming that the mass number is about $2Z$. For atoms knocked-on by charged particles, on the other hand, the condition $Z > 10$ usually suffices.

Electronic excitation.—Fast charged particles produced by accelerators lose most of their energy by electronic excitation in the initial part of the range. Over most of their range the energy lost to atomic collisions is only about 10^{-3} the loss to electronic excitation. By contrast, knocked-on atoms are almost always so slow that appreciable electronic excitation cannot occur and a theory based entirely on atomic encounters is adequate.

In the transition range, both theory and experiment give inadequate information about the electronic loss, so that considerable uncertainty exists in such cases as to the total fraction of the initial kinetic energy of the particle which is expended in the production of displaced atoms. In the case of fission products, it has been estimated that about 9 per cent of the total kinetic energy of a fragment is expended in atomic collisions, i.e., either displaced atoms or local thermal excitation of the lattice (10). In this case, Ozeroff (10) shows that the difference between the classical and Born approximations for the primary collisions is of significance, the average knock-on energy being 1900 ev in the classical case compared to 375 ev in the Born approximation.

Lassen (11) has described interesting studies on the energy losses of fission fragments passing through gases and solids. He has developed elegant experimental methods for measuring the range-energy curves in detail and also for evaluating the average charge of a fission fragment along its path. For fragments in uranium, the average charge in the initial part of the range is 22 for the heavy fragment and 20 for the light fragment, somewhat less than theoretical estimates.

In the case of light elements, electron excitation must be considered for charged particle knock-ons for $Z<10$, and in the case of fission neutron knock-ons for $Z<15$. This is because the knock-ons from neutron collisions are distributed uniformly in energy, whereas for the Rutherford type collisions of charged particles, the differential cross section for energy transfer T is proportional to dT/T^2, and is thus weighted strongly towards low energies. Roughly speaking, electronic excitation becomes small when the velocity of the particle is small compared with the velocity of the slowest electrons in the atom, or in the case of metals, with the velocity of electrons at the Fermi surface. In the case of insulators, Seitz suggests (9) that the electron energy corresponding to the velocity of the moving atom should be less than one-fourth the energy gap. For the average carbon knock-on from fission neutrons in graphite, the equivalent electron energy is about 13 ev, showing that substantial energy must be dissipated in electron excitation.

Damage by electrons and gamma rays.—Recently, fast electrons and gamma radiation have been used increasingly to generate radiation damage. Electrons of energy sufficient to produce displacements are in the relativistic range. Even for energies considerably above threshold the cross section for a displacement encounter is hence about 100 times smaller for an electron than for a charged nucleon. Furthermore, since the collisions are heavily biased towards small energy transfer, the knocked-on atoms seldom have energy to produce secondaries, and in consequence electrons tend to produce a simpler type of damage than other particles, namely, isolated pairs of vacancies and interstitials. On the other hand, the range of electrons is usually short so that their capacity for producing damage varies appreciably through the depth of the sample, especially if their energy is near threshold. This inhomogeneity in depth is often inconvenient for making interpretable measurements of properties.

Recently Dugdale (14) has pointed out that gamma rays may have sufficient energy so that some of the Compton and photoelectrons generated by them exceed the threshold for displacement production. Dugdale showed that disordering could be produced in Cu_3Au by means of Co gamma radiation. More recently Cleland *et al.* (15) have used Co gamma radiation to produce displaced atoms in Ge. This type of irradiation probably produces the simplest type of defect structure possible. The energies of the electrons are close enough to threshold that only single pairs of vacancies and interstititals are generated, and the absorption of the gamma radiation is small enough to produce a much more uniform distribution in depth than electrons of comparable energy. The displacement efficiency per photon is about 10^{-3} that of fast neutrons in the case of Ge (15).

Measurements of threshold energy.—Electron bombardment is especially convenient for measuring the threshold energy for displacement production. This has been done most accurately in the case of Cu and Ge. The value of Cu is given as 25 ev (13) and that for Ge as 31 ev (12). Recently attempts have been made to calculate these displacement energies theoretically.

Huntington (16) has considered the low energy collisions in Cu using a Born-Mayer type repulsive potential between ions deduced from elastic constants. For knock-ons in various directions and for two limiting assumptions about the potential, the values of E_d range between 17 and 43 ev, bracketing the observed value. On the other hand, a similar calculation by Kohn (17) for Ge leads to a value of only 10 ev, which is much smaller than observed. It is impossible to say at present whether the discrepancy is due to simplifying assumptions made by Kohn in the treatment of interatomic forces, or whether, as Kohn suggests, the interstitital position assumed in his calculations is not stable. It may also be remarked that the experimental situation is not entirely clear. Recently, Loferski & Rappaport have made a determination of the electron bombardment threshold for Ge and Si based on the reduction in minority carrier lifetime, which is a considerably more sensitive indicator than resistivity (19, 20). They conclude that the displacement energy for both Ge and Si should be taken as 13 ev, in much better agreement with the calculations of Kohn. However, it should be pointed out that the appearance of the damage near threshold is very gradual (20) indicating that the threshold is actually quite spread out in accordance with the ideas of Sampson *et al.* (34). Thus, the displacement energy used in calculating the number of displacements should still probably be the higher value of Klontz (12).

There are two other direct measurements of displacement energy. One is due to Eggen (106) who found a value of 25 ev for graphite by electron bombardment. The other is due to Denney (21), who measured the threshold for the conversion of the paramagnetic Fe precipitate in 2.4 per cent Fe-Cu alloy to the ferromagnetic form. The value $E_d = 27$ ev which was found by electron bombardment was originally attributed to Fe, but detailed consideration of the mechanism of the induced precipitation suggests that it is due to defects generated in the Cu matrix, and the E_d value should thus be 24 ev for Cu. It would be interesting to carry out displacement measurements on Al, which is similar in structure to Cu, but should have a much smaller ion core repulsion.

Formulas for total number of displacements.—Much attention has been devoted recently to computing the total number of displaced atoms (3, 9, 22, 23). The calculations are made on the assumptions that (*a*) the collisions are all binary and of the hard sphere type, (*b*) the atoms of the solid may be treated as randomly distributed, and (*c*) each lattice atom is bound to its normal position in a square potential well of depth E_d. With these assumptions it is possible to solve rigorously for the number of displaced atoms. For a primary energy E', the result for the total number of displacements including the primary is approximately (9, 23):

$$\nu(E') = 0.561\left(1 + \frac{E'}{E_d}\right) \qquad 2.$$

This is an excellent approximation provided $E' > 4E_d$ (9). The linearity in

E' implies that for most cases the total number of displacements can be calculated in terms of the total energy which is expended in atomic collisions. Equation 2 indicates that roughly half the total energy goes into the production of displaced atoms, and the other half into excitation of lattice vibrations. Similar conclusions have been reached by Snyder & Neufeld (22) and by Kinchin & Pease (3), except that the fraction is exactly 0.5 instead of 0.561. For charged particle bombardment, the distribution of the primary energies E' is heavily weighted towards low energies, and the corrections to equation 2 for low energies are of some importance. The expression for the average number of displacements per primary collision becomes (9) (23):

$$\bar{\nu} = 0.107 + 0.561 \ln\left(1 + \frac{E_m}{E_d}\right) \qquad 3.$$

In equations 2 and 3 E_m is the maximum energy which can be imparted to a primary particle by the bombarding particle, i.e.:

$$E_m = \frac{4M_1M_2}{(M_1 + M_2)^2} E' \qquad 4.$$

where M_1 and M_2 are the masses of bombarding and lattice particles respectively. In cases where electronic excitation is important, equation 4 should be replaced by the energy for which electronic excitation becomes significant. For the case of 12 Mev deuterons on Cu, equation 3 gives a value $\bar{\nu} = 6.3$, which is about twice the estimate of the earlier theory (8). The number of primaries for track length dR of the bombarding particle is:

$$dn_p = N_0 \frac{\eta}{E'} dR \qquad 5.$$

where N_0 is the number of target atoms per unit volume, E is the incident particle energy, and η is given by:

$$\eta = 4\pi a_h^2 \frac{M_1}{M_2} \frac{Z_1^2 Z_2^2 R_h^2}{E_d} \qquad 6.$$

where a_h is the Bohr radius and the other symbols are as previously defined. These formulas give, for example, about 3×10^4 displaced Cu atoms per cm path length of a 12 Mev deuteron, or 600 displaced atoms in the total range if the deuteron is stopped in the target. For fission fragments having a total kinetic energy of 160 Mev, about 14.5 Mev (10) is expended in atomic collisions, which gives 325,000 displaced atoms per fission, or 160,000 per fragment, assuming $E_d = 25$ ev for uranium.

For neutron collisions the analogue of equation 4 is:

$$\bar{\nu} = 0.56 + 0.28 \frac{E_m}{E_d}$$

which gives about 1370 displacements per average fission neutron collision

in copper. For the case of graphite, taking as 3 ev the energy below which electronic excitation becomes negligible for the equivalent electron energy, or 6.6×10^4 ev, the number of displacements is only 740 per primary neutron collision, and becomes insensitive to neutron energy for neutrons above about 0.23 Mev.

Seitz & Koehler (9) have given arguments to show that equation 2 may be used even when the hard sphere assumption is invalid and the collisions are of the Rutherford type. Thus the application of these results to graphite and to neutrons and fission product damage, as above, is insensitive to this assumption.

Experimental verification of the displacement theory.—Several attempts have been made to obtain an experimental check on the theory for the total number of displaced atoms. These are: (*a*) A measurement of the diffuse scattering of long wavelength neutrons in irradiated graphite (24). (*b*) An estimate of the number of Frenkel pairs produced in Cu by deuteron bombardment at 10°K. deduced from the residual electrical resistivity of the bombarded material (6). (*c*) An estimate similar to (*b*) for Cu bombarded with 0.81 Mev electrons at a temperature between 85°K. and 125°K. (13). (*d*) A measurement of the initial rate of loss of conductivity of *n*-type Ge bombarded with 1.5 Mev electrons (25) and with 4.5 Mev electrons (26) at liquid nitrogen temperature. (*e*) A measurement of the release on warming to room temperature of the energy stored in Cu bombarded at liquid N_2 temperature. All of these measurements except (*a*) have been discussed at length (9), and so we shall make only a few summarizing comments here.

Method (*a*) can be used for solids in which the diffuse scattering background due to thermal agitation, random distribution of isotopes, and random distribution of nuclear spins is sufficiently small (24, 28). If the wavelength is long enough so that Bragg reflection cannot occur, this is a powerful tool, since in contrast with conduction electrons, the laws of scattering of neutrons from interstitials and vacancies are precisely known. Graphite is a particularly favorable example because its Debye temperature is high, and it consists almost entirely of a single isotope of spin 0. In the Brookhaven experiment the transmission of an unirradiated sample was compared with that of a sample irradiated at room temperature to approximately 1.1×10^{20} neutrons/cm^2. In order to correct for spurious intensity changes attributable to the change in *c*-axis spacing of irradiated graphite, the transmission was measured as a function of wavelength with the aid of a crystal spectrometer and the change in integrated intensity thus estimated. The fraction of displaced atoms was found to be 2.6 per cent with an error of about 9 per cent. A theoretical calculation yields at least twice this value. The computation, however, contains many uncertainties not only because of the inaccuracy in the estimation of the energy lost to conduction and valence electrons, but also because of uncertainties in the magnitude and energy distribution of *nvt*. Together these could easily amount to an error of a factor of more than 2. The technique would appear very promising as a comparative calibration method

for neutron fluxes used in other radiation damage experiments, as it automatically integrates over the energy distribution of the neutrons.

Methods (*b*) and (*c*) unfortunately depend upon theoretical calculations of the scattering of conduction electrons by vacancies and interstitials. Such calculations have been made by Jongengerger (29) for vacancies in Cu and by Blatt (30) for interstitials. The scattering calculations were made by the method of partial waves, treating the conduction electrons as free and using self-consistent potentials for the defects. The results were found to be fairly insensitive to the form of the potential, provided it satisfied the Friedel sum rule (31), which is an expression of the conservation of charge. On the other hand, more recently Blatt *et al.* (32) have used the same methods to compute the resistivity due to Ga, Ge and As impurities in Cu, and have found values which are too high by a factor of 2, suggesting that the resistivity of imperfections may have been similarly overestimated. At all events, the calculations give a resistivity of 2.7×10^{-6} ohm cm. for each 1 per cent Frenkel defects in Cu and this when combined with the theoretical estimate of the number of pairs produced by the bombardment, gives a value of the residual resistivity which is higher than observed by a factor of 5 for deuteron bombardment at 10°K. and by a factor of 7.5 for bombardment at 80°K. In the case of electron bombardment the discrepancy is about 5.7 for bombardment at 80°K. (9, 13). It is interesting to note that Blewitt and coworkers (33) have recently found a similar discrepancy of a factor of 5 with fast neutron bombardment at about 22° in the Oak Ridge reactor.

Sampson, Hurwitz & Clancy have pointed out that the theoretical estimate of the number of displacements is sensitive to the assumed form of the probability of displacement as a function of energy (34). As we have seen, the usual theory assumes that the probability rises abruptly to unity at the threshold energy E_d. Sampson *et al.* show that if a linear variation between E_d and a higher energy E_1 is assumed, the number of displacements depends more on E_1 than on E_d. This could considerably reduce the theoretical number.

Cooper and coworkers (6) have compared the relative resistivity changes produced in Cu, Ag, and Au with the theory. Equations 5 and 6 show that the number of displacements should be proportional to $(Z^2N_0/M)\bar{\nu}$ where the constants pertain to the target atoms. The damage for Cu, Ag and Au should thus be in the ratio 1.00:1.025:1.50, as compared with an observed ratio 1.00:1.17:1.70. The agreement is fairly good and makes the disagreement in absolute magnitude difficult to account for.

Method (*d*) should be the most reliable available at present, since it is less dependent on theory. The work of Klontz (25, 26) at liquid nitrogen temperature showed that in *n*-type Ge, 0.065 carriers were lost per bombarding 1.5 Mev electron, and the ratio was 6.15 times higher for 4.5 Mev electrons as compared with a theoretical factor of increase of only 2.6 (9). This difference is in accord with the suggestion of Sampson *et al.* (34). Recent work at Oak Ridge (35) confirms that two types of electron traps are

produced by both electron and neutron bombardment in Ge. This is compatible with the model suggested by James & Lark-Horovitz (36), provided we assume that the two traps are associated with a Frenkel pair. On this hypothesis, the 1.5 Mev bombardment produced 0.032 displacements per electron and the 4.5 Mev bombardment 0.196 displacements per electron compared with theoretical values (9) of 0.06 and 0.156 respectively. For the experiments of Fletcher, Brown & Wright with 3 Mev electrons at room temperature, the theoretical estimate for the number of displacements is about 4 times the observed, but in this case some annealing undoubtedly occurred (37). For alpha bombardment at room temperature, the theoretical number of displacements is about 3 times the observed (38, 39, 9). A discrepancy of a factor of 4 is found for deuteron bombardment at dry ice temperature (39, 9). All the above figures are based on the assumption of two acceptors per displacement, and hence the estimates differ by a factor of 2 from those given by Seitz & Koehler (9).

An estimate of the number of traps produced can also be made on the basis of the deuteron bombardment required to produce conversion from *n*- to *p*-type Ge at 90°K (40). The evidence is somewhat conflicting, but for low initial carrier concentrations, it appears that the observed number of defects is too small by a factor of approximately 8.5. For high initial carrier concentrations, the discrepancy is only a factor of 1.6, but here the data are less reliable because of clustering and radiation annealing effects which may take place at high bombardments. The factor of 8.5 probably represents the most reliable experimental result since it does not depend on assumptions regarding carrier mobility. For neutron bombardment at liquid nitrogen temperature, the Oak Ridge group (35) has found an average of 5 acceptor levels per cc. per unit of fast effective *nvt*. If 60 per cent of this effect is ascribed to mobility, as will be shown later, this corresponds to only 1 Frenkel pair per *nvt*, a value which is approximately 15 times too small. The larger discrepancy in this case may arise from the fact that neutrons tend to produce clusters of displacements separated by relatively less disturbed material (see below). The conclusion is thus not too different from that arrived at in the case of Cu.

Method (*e*) has been carried out by Overhauser (27), who computed the number of vacancy-interstitial pairs in his samples on the basis of Huntington's most recent estimate of 5 ev for the energy of formation of a Frenkel defect in Cu (41). He found that a resistivity of 11 ohm cm. per 1 per cent Frenkel pairs would be necessary to reconcile the electrical results of Cooper *et al.* (6) with the stored energy measurements. This would make the number of Frenkel pairs 20 times smaller than theory for Cu. Overhauser has suggested (42) that the discrepancy with Blatt's resistivity calculations can be resolved by taking into account the very large scattering which results from lattice strain around an interstitial, and which was neglected by Blatt (30). The discrepancy with theory he believes can be accounted for by ideas similar to those of Sampson *et al.* (34). It would be interesting to carry out stored

energy measurements in Ge or Si subjected to electron bombardment, where interpretation of the results would be subject to less ambiguity. It is to be noted also that Overhauser's speculation is consistent with the density measurements of Kierstead (172).

Mean free path between collisions.—It is interesting to estimate the mean free path between the displacements produced by a primary knock-on. This has been discussed by Ozeroff (10) and by Brinkman (43) on the basis of an assumed repulsive potential between colliding atoms of the form:

$$v(r) = \frac{Z_1 Z_2 e^2}{r} e^{-r/a} \qquad 8.$$

Here Z_1 and Z_2 are the atomic numbers of incident and target atoms respectively, while a is given by the relation:

$$a = \frac{\lambda a_h}{Z_2^{1/3}} \qquad 9.$$

Brinkman and Ozeroff took the parameter λ as 2.09 based on the Thomas Fermi model whereas Seitz and Koehler (9) recommend a value of 0.71. The latter value gives $a = 0.125$ Å for Cu. From the elastic constants Huntington has bracketed a between 0.15 and 0.195 Å (44) corresponding to λ between 0.85 and 1.10. For energies normally encountered the collision radius for energy transfers exceeding T is given to a good approximation by the solution of the equation:

$$R = a \ln \sqrt{\frac{\pi}{2}} \frac{Z_2^2 e^2}{\sqrt{aR}\sqrt{TE'}} \qquad 10.$$

where E' is the energy of the incident atom. The corresponding mean free path between collisions which transfer energy T is:

$$L_T = \frac{4}{3} \frac{r_s^2}{R^2} \qquad 11.$$

For $T = E_d = 25$ ev, $E' = 300$ ev, characteristic of average conditions for 12 Mev deuterons on Cu, L_d is 6.2 Å. On the other hand for the same T and $E' = 10^6$ ev, characteristic of the maximum energy primaries for fission neutron bombardment of Cu, L_d becomes 17.7 Å, the latter representing about 6 or 7 atom spacings. For collisions in which the atom loses more than $\frac{1}{4}$ its initial energy, the mean free path is about 150 Å.

The estimates of the preceding paragraph give us a rather different picture of the spatial distribution of the damage associated with a single primary collision. Especially for neutron damage, the energy is dissipated over quite a large volume, the estimated mean free paths being about 10 times larger than those derived in earlier theories (43). For fast neutrons the energy of the primary knock-on may well be communicated to a volume containing of the order of 10^6 atoms, and as will be seen this means that the initial amplitude of the associated "thermal spike" is not as large as was originally believed. It should also be mentioned that the scattering will be

somewhat preferentially forward in the laboratory system, so that the energy will tend to be dissipated in a spheroidal rather than a spherical region, more extended in the original knock-on direction. The damage produced by neutrons tends to be quite inhomogeneously distributed, and a rough estimate indicates that a fast neutron *nvt* of 10^{18} would be required to produce significant overlapping of the damaged regions. The situation is much more favorable for charged particles, as an irradiation of 10^{15} particles per cm^2 or 10^{-7} ampere-hr. per $cm.^2$ is sufficient to produce displacements which are uniformly distributed.

Evidence for nonuniformity of neutron damage can be drawn from the experiments of the Oak Ridge group on Ge (35, 45). The non-random distribution of electron and hole traps associated with the damaged regions results in large fluctuations in the potential in the semiconductor interior. The fluctuations give rise to an anomalous dependence of mobility on temperature, much more rapid than for the normal mobility resulting from ionized impurity scattering. The origin of this effect is probably somewhat analogous to that of the high dislocation scattering in *n*-type Ge as discussed by Read (46). The inhomogeneity of damage, which becomes more serious for heavier elements, is a serious complication in the interpretation of neutron damage experiments, but is sometimes overlooked. A similar problem occurs in connection with fission damage.

Secondary Effects of Collisions

Disordering by radiation.—As we have seen, about half the energy expended in collisions with the lattice atoms actually goes into encounters in which energy less than E_d is transferred, in other words, collisions without displacements. These low energy collisions give rise to a number of secondary effects, which have been the subject of considerable speculation. The motivation for this speculation is the observation that radiation can produce atomic rearrangements in metals which are more extensive than can be accounted for by displaced atoms alone. For example, Aronin (47) has studied the disordering of Ni_3Mn by fast neutron bombardment, and has estimated that over 5000 atoms are disordered for each collision by a neutron over 0.5 Mev, whereas not more than 1300 displacements could have been produced. Disordering was also observed in Cu_3Au, originally by Siegel (48). Quantitative estimates of the number of disordered atoms per primary knock-on can be made from more recent work. For 9 Mev proton bombardment (49) and 33 Mev alpha bombardment (50) the number of disordered atoms per primary has been estimated at 125. This is to be compared with values of $\bar{\nu}$ of 5.7 and 12.7 for the Cu atoms, respectively. Comparable results have been obtained for beta brass (51).

Thermal spikes.—Several theoretical models have been brought forward to account for these large disordering effects. The simplest in conception and the oldest historically is the thermal spike idea, suggested first in this connection by Seitz and extended by Brooks in unpublished work during and

after the war (52). Our present picture of the thermal spike must be somewhat modified, however, in the light of the longer mean free path for collisions discussed above. Originally the picture was that the primary displaced atom produced very intense heating in a region of atomic dimensions, and the resultant energy then diffused outwards according to the laws of heat conduction in solids. Thus the temperature behavior at a distance r and time t from the origin of the spike would be:

$$T(r, t) = \frac{Q}{\rho c} \frac{1}{[4\pi D(t + t_0)]^{3/2}} \exp\left[-r^2/4D(t + t_0)\right] \qquad 12.$$

Here Q is the energy of the primary knock-on, ρc the heat capacity per unit volume of the lattice, D the thermal diffusivity, and t_0 a parameter which defines the volume over with the original heat is developed. In metals the thermal conduction is normally by electrons, but the duration of a thermal spike is so short, about 10^{-11} sec., that no exchange of energy between electrons and lattice can occur (52). Thus the thermal diffusivity which appears in 12 should contain only the lattice contribution to the conductivity and is probably only of the order of 0.001 cm.²/sec. (9). As noted previously, the mean free path of a primary knock-on is of the order of 7 to 18 Å and in consequence Seitz & Koehler point out that Q is actually spread out in several small hot spots separated by a distance of this order. After the heat from these spots merges, equation 12 is fairly well applicable, but one should accordingly take $t_0 = r_0^2/4D = 100\ r_s^2/4D = 0.5 \times 10^{-11}$ sec. and the initial temperature at the maximum as about 1300°C. above ambient for a 500 ev knock-on in Cu. For a fission neutron knock-on, the mean temperature reached may actually be of the same order of magnitude since the mean free path of the primary knock-on is longer, and the initial energy thus distributed in a larger volume. However, considerably higher temperatures may be reached locally at the point of generation of secondaries.

A point of view which is often adopted is to compute the number of activated reactions which can occur in a thermal spike when the activation energy is E'. The result is (9):

$$n_T = 0.016\, \rho' \left(\frac{Q}{E'}\right)^{5/3} \qquad 13.$$

where

$$\rho' = \eta \frac{\nu_0 r_s^2}{D} \qquad 14.$$

In the second equation ν_0 is the frequency of oscillation of an atom, r_s is the radius corresponding to the atomic volume, and η is the number of independent paths by which the given reaction may take place. Equation 13 ignores ambient temperature and also ignores the t_0 effect mentioned above. The first leads to an underestimate, the second to an overestimate of the number of transitions n_T. In general, ambient temperature effects are of no importance unless the process is one which would almost but not quite go at

ambient temperature anyway. Taking ambient into account leads to radiation-induced processes which are temperature dependent (52).

The result of equation 13 is very sensitive to the quantity ρ', which is related to the ratio of the diffusivity for atoms to the diffusivity for heat. It is probably close to 1, and more likely less than more. For the disordering of Cu_3Au by deuterons, Seitz & Koehler (9) take the thermal spike as made up of 6 separate spikes of 50 ev each and conclude from 13 that the number of disordered atoms is about 10 for $E' = 3$ ev and $\rho' = 1$. This is much smaller than the 125 observed for the computed conditions. It is difficult to make a sensible calculation for neutrons for comparable assumptions, since the initial energy is distributed spatially in a complicated way. A more precise solution of the spatial slowing down problem, treating the lattice as a gas, might actually be of some value in this connection, but this has not been carried out.

Plasticity spike.—The conclusion is that it seems unlikely that the thermal spike mechanism can account for the observed disordering. As an alternative, Seitz has suggested (53) that the local plastic formation around a thermal spike results in disordering. It is necessary that the resultant shears be highly irreversible, and Seitz suggests that this arises from mutual entanglement of the small dislocation loops produced on different slip systems around the spike. While it appears that this mechanism could account for the observed effect, it is difficult to use for quantitative calculations.

Replacement collisions.—Kinchin & Pease (54) have suggested an alternative mechanism which has the attractiveness of being both simple and subject to quantitative predictions. They point out that a moving atom which does not have enough energy to displace another atom may nevertheless have enough energy to substitute for it and push it into an interstitial position. This mechanism, termed a replacement collision, will not increase the total number of displaced atoms but can substitute a wrong atom for a right atom in an ordered alloy. It would also serve as a mechanism for dispersal of interstitials in a pure metal. Kinchin & Pease give a simple equation for the ratio replacement to displacement collisions

$$\frac{R}{D} = \frac{1}{2}\left[1 + \frac{\ln (E_d/E_r)}{\ln 4/3}\right] \qquad 15.$$

where R is the number of replacements, D the number of displacements, and E_d and E_r the corresponding energies. The energy required for a wrong atom to replace a right atom is considerably more than the energy required for a replacement of similar atoms. Kinchin and Pease take it as 2.5 ev for Ni_3Mn and thus account for 6100 replacements in Aronin's (47) experiments. They thus account for 7400 events of which 3/8 or 2800 are disordering events. Considering all the uncertainties the agreement is not bad. On the other hand, equation 15 does not give a sufficient ratio to account for the results of charged particle bombardment of $AuCu_3$, unless one assumes unreasonably small values for E_r. It would be valuable to carry out detailed calculations

on the replacement mechanism similar to those of Huntington on Cu (16) for the displacement mechanism.

The work of Cook and Cushing on the neutron bombardment of Cu_3Au (55) and CuAu (56) should also be mentioned. Unfortunately, their flux estimates were not very accurate, but they conclude that one neutron collision disorders about 10,000 atoms. This number could be accounted for by thermal spikes only if it is assumed that the initial heat due to the collision is considerably more concentrated than indicated by the discussion above. The replacement collision mechanism also appears to fall short of explaining the results. Careful work showed in this case that ambient temperature effects were not present.

Phase changes under irradiation.—There has been considerable interest in the problem of the promotion of phase changes by irradiation. Three different types of effects might be expected. (*a*) A thermal spike could result in the formation of a nucleus for a new phase which would subsequently grow on warming. (*b*) The production of imperfections could result in accelerated growth of a stable phase for which nuclei are already present. (*c*) Radiation could result in the resolution of a precipitate either through atomic displacements or through a thermal spike effect. Examples of all of these effects have been observed, but they do not always occur, and the favorable conditions are not too clearly established.

Perhaps the most dramatic example of the first effect is the work of Fleeman & Dienes on grey tin (57). Whereas unirradiated samples, supercooled and then held at −50.3°C., showed negligible rate of transformation to the α-phase, similar samples irradiated to 10^{18} *nvt* of fast neutrons at 80°K., and then warmed to −50.3°C., transformed in about 1300 minutes. The irradiated samples showed a longer induction period and a slower rate of transformation than seeded samples at the same temperature. This fact suggests that the neutron effect was really to promote nucleation and not just to accelerate growth by providing defects. That the effect is possibly attributable to thermal spikes is further indicated by the fact that no transformation is observed as a result of electron bombardment (58). It would be interesting to study the effect with bombardment by other charged particles of varying energies in order to estimate the magnitude of the total energy transfer required to create a nucleus.

In contrast to the grey tin case, Reynolds, Low & Sullivan (59) have investigated the stability of type 347 Stainless Steel under fast neutron bombardment. This is an austenitic steel which is metastable at room temperature with respect to the ferritic phase. Some conversion was observed, but the amount was proportional to the amount of ferrite already present as a result of cold work. Very similar results are reported by Konobeevsky *et al.* (60). It should be stressed, however, that this experiment is not strictly comparable to the one on tin because the irradiation was carried out relatively closer to the transition temperature. An important factor in the tin

experiment may lie in the fact that the driving free energy at the irradiation temperature was large and that hence the critical size nucleus may have been small enough to form within a thermal spike. It would be interesting to carry out the tin experiment by irradiation at a higher temperature, and the steel experiment with irradiation at a lower temperature to test this hypothesis. In the experiments of Reynolds *et al.* it seems clear that the effect of the irradiation was merely to promote growth of a phase already present. Somewhat contrary to this, Denney (61) has found that the austenitic precipitate of Fe in a Cu 2.4 per cent Fe alloy is actually metastable and can be converted to ferrite by 9 Mev proton bombardment. Apparently, this result is a function of the degree of metastability, i.e., of the size of the critical nucleus under the conditions of irradiation.

Another experiment of the same type has been reported by Tucker & Senio (62). They exposed a metastable U 2 per cent Cr alloy in a reactor and failed to observe growth of a second phase. They interpreted their results as throwing doubt on the reality of thermal spikes, but there are too many other factors involved to warrant such a definite conclusion.

A system which has been studied extensively is the supersaturated solid solution BeCu. Murray & Taylor suggest (63) that neutron irradiation of this alloy at room temperature results in the formation of a small precipitate equivalent to that which forms on aging at about 100°C. Furthermore, by low temperature irradiation (64) it can be shown that this precipitate is "latent," that is, it does not appear until the specimen is reheated to room temperature, indicating that nucleation must take place, followed by growth attributable to additional defects present when the alloy is warmed. If the sample is bombarded at 120°K. and then given additional bombardment at room temperature, the change in resistivity is the same as though the whole bombardment had been carried out at the higher temperature, indicating that all the defects are stored. Presumably the original nucleus must have been due to a thermal spike as in the case of grey tin.

The picture which emerges from the BeCu work is further strengthened by work on the alloy BeNi, which has the advantage that the amount of precipitation can be determined quantitatively from the Curie temperature of the ferromagnetic solid solution. A solution of 14.65 atom per cent Be in Ni was irradiated to an *nvt* of 4×10^{17} neutrons/cm.^{2} at 300°C. (65). It was found 1.1×10^{21} Be atoms were precipitated by the radiation, or about 30 per displaced atom. The work of Cleland *et al.* (64) on BeCu shows that none of the enhanced precipitation can be due to thermal spikes. Therefore it must be concluded that the excess vacancies produced by the irradiation must make many more jumps before being annihilated than the average Be atom does in precipitating!

A case in which irradiation apparently produces a transformation against the direction favored by thermodynamics has been reported by Konobeevsky *et al.* (66). They found that in a stable two phase system of a U 9 per cent Mo

alloy, fission product damage dispersed the precipitated phase and rendered the alloy homogeneous. The metastable single phase alloy showed no precipitation.

The disordering of ordered alloys is, of course, another example of a reaction under bombardment which reverses thermodynamics. It is also reported that the slight short range order existing in α-brass is removed by irradiation (67).

The direction of the reaction produced by irradiation is evidently a sensitive function of temperature. This is well illustrated in the case of the system Cu_3Au. Coltman & Blewitt (68) first reported that the ordering rate of this alloy could be increased by prior irradiation of quenched disordered samples at room temperature. They interpreted this as evidence of diffusion enhanced by the presence of extra vacancies introduced by the irradiation. A quenched alloy annealed at 150°C., well below the ordering temperature, showed no ordering when held at temperature, but ordered rapidly when irradiated to only 2.5×10^{13} neutrons/cm.^{2} at the same temperature. On the other hand, when irradiated at -160°C., only a slight increase in resistivity from introduced defects was observed (69). This shows that the temperature either during or after irradiation must be high enough so that the defects migrate. Analogous effects were found by Brinkman (70) in an alloy quenched from close to the melting point in order to freeze in vacancies. Dixon & Bowen (71) irradiated ordered Cu_3Au with 36 Mev α's at -180°C. and $+220$°C. and found that the disordering rate at the higher temperature was 6 times smaller than at the lower. They interpreted this result as indicating that ordering and disordering were occurring simultaneously. Irradiation with electrons just above the threshold for displacing Cu results in no observable disordering, but still generates sufficient defects to induce ordering when the specimens are subsequently heated to 130°C. (70, 72). Similar effects have been observed with gamma rays (14).

The phenomenon of radiation annealing (73) constitutes evidence for the reality of a thermal spike mechanism. In general, it is found that reactions go in the presence of radiation which would be totally inhibited in the absence of radiation at the same temperature. Such an effect cannot be ascribed unequivocally to thermal spikes, however, even when it is temperature dependent. A simple example is given in the work of Cooper *et al.* (6). There was a definite difference in the saturation of the deuteron bombardment damage for irradiation of Cu at 10°K. and 16°K., which could not be ascribed to purely thermal recovery at the higher temperature. The saturation was also much greater than would be explained by displacement of already displaced atoms, or even by the replacement collision mechanism. It would be most interesting to carry out comparative studies of neutron, deuteron, and electron damage versus bombardment time at carefully controlled temperatures and with comparable amounts of total damage. The tendency towards saturation should show marked differences in the three cases, being almost totally absent for electrons, if it is due to thermal spikes. If higher order reac-

tions are involved, radiation annealing will be further enhanced by inhomogeneity in the damage distribution. It is only necessary that the recovery involve thermally activated migration of a defect which results from the primary displacement process. An effect of this sort has been pointed out by Marx (74) in another connection.

Other evidence for radiation annealing is discussed in the section on graphite.

Brooks (52) has given a quantitative discussion of the influence of ambient temperature on the enhancement of activated processes in the presence of radiation. Chang (75) has given a nice physical picture of the process, pointing out that when the ambient temperature is such that the process is almost able to take place thermally, very large radiation enhancement can result because the temperature is raised even in the long decay tail of the spike so that significant fractions of the total volume at any given time are activated.

Damage mechanisms in nonmetals.—In metals it has been noted that gamma and beta radiation cannot produce damage unless the particles have sufficient energy and momentum to give a recoil energy of the order of 25 ev to a lattice atom. Seitz & Brooks (9, 52) have shown by detailed arguments that energy which goes into ionization is dissipated within the electronic system without appreciable transfer to the lattice. This is because the electron-lattice interaction is weak. There is a possibility that *d*-electrons in transition metals or *f*-electrons in rare earths might exchange enough energy with the lattice to produce observable effects, but this suggestion has never been tested experimentally (9).

The situation is quite different in nonmetals, however. There is ample evidence to indicate that displaced atoms in the form of vacancies can be produced by soft x-rays and ultraviolet radiation in alkali halides (76) even at liquid He temperatures. The criterion for this is that the excited electronic states of the solid, i.e., the first forbidden energy gap, should have an energy of excitation comparable with that needed to produce a displacement. It is often convenient to describe the damage to insulators in terms of the rupture of bonds, which allow the atoms to recoil with large momentum.

In ionic crystals Seitz (77) suggests that vacancies can be generated at jogs in dislocations as the result of capture of excited electrons at "incipient vacancies." Recently Dexter (78) has given a more quantitative discussion of this mechanism, and finds some difficulties, perhaps not insurmountable, in the detailed interpretation of the experiments. In particular, the experimental fact that the production rate is nearly independent of the concentration of *F*-centers is rather surprising on this model. Recently Varley (79) has suggested an alternative mechanism which is simple and ingenious. Consider the NaCl lattice and suppose that through electronic excitation the Cl^- loses two electrons to become Cl^+. The positive ion will then find itself with an electrostatic energy which is of the order of twice the binding energy of Cl^- in the crystal, and will therefore seek either an interstitial site or a vacant

Na^+ site, provided it can move before it recaptures an electron. Varley attempts to explain all the color centers on the ultraviolet side of the F-band in terms of defects generated by this mechanism. No detailed theoretical analysis of the displacement process has been given, and there may be some question whether the lifetime of the Cl^+ against capture of an inner electron from an adjacent Na^+ is sufficiently long to permit a displacement to be completed. The process suggested by Varley has the property that it is not structure sensitive, i.e., it would not depend on the presence of dislocations or other prior crystal imperfections. As shall be seen there is in fact evidence for both structure-sensitive and non structure-sensitive processes in the coloring of alkali halides.

Other damage mechanisms.—Smoluchowski has recently started radiation damage work with very high energy particles (~300 Mev) from the cyclotron of the Carnegie Institute of Technology. These fast particles have substantially lower collision probability for displacement collisions than do the particles in the 10 Mev range used by other groups. Furthermore, they have a high probability of producing disruptive nuclear collisions in which several energetic charged particles are emitted. Smoluchowski shows (80) that the damage produced by nuclear reactions may actually exceed the damage produced as a result of primary knock-ons.

Damage can also be produced as a result of transmutation reactions with thermal neutrons. Two different mechanisms are involved. In the first place, the nuclear transmutation introduces a chemical impurity which may influence the physical properties of the crystal. For example, Cook & Cushing (55, 56) have shown that thermal neutrons alone can enhance the ordering rate of Cu_3Au and CuAu. They ascribe this effect to the presence of Hg produced by transmutation of Au. Cleland & Crawford (81) have observed changes in the electrical properties of InSb resulting from the transmutation of In to Sn by neutron capture. Similar effects had been observed much earlier by the Purdue group in Ge and Si. In the second place the capture gamma ray released in the reaction gives sufficient recoil energy to the atom to displace it from its normal lattice position, so that the transmuted impurity usually appears in an interstitial position and leaves a vacancy in the normal lattice. This could also have produced the effects observed by Cook & Cushing.

Still another damage mechanism which should be mentioned occurs in compounds of B and Li, of which boron nitride (81a) and lithium fluoride (201, 202, 203) are the most extensively studied. In these cases the capture of a thermal neutron results in the emission of an energetic alpha particle which is responsible for more displacement damage than the accompanying fast neutron flux.

REACTOR MATERIALS

Graphite

Physical properties and electronic structure.—Graphite, as used in reactors,

is an exceedingly complex material. It is made by partial recrystallization of carbon residues or coke at extremely high temperatures. A comprehensive review of the physical properties and manufacturing processes for reactor graphite has been given by Currie *et al.* at the Geneva conference, and the reader is referred to their paper for details (82) and for a fairly complete bibliography.

Graphite crystals have a somewhat unusual electronic structure originating in their highly anisotropic crystal structure. The crystal consists of tightly bonded planes with covalent binding, loosely stacked and held together by Van der Waals forces (83). The electrical resistivity perpendicular to the planes is of the order of 100 to 1000 times greater than that in the planes, this property being in all probability rather structure sensitive (84, 85). The resistivity is 5×10^{-5} ohm/cm. at room temperature and has a strong positive temperature coefficient. The resistivity of natural graphite becomes extremely low at very low temperatures, while for fabricated graphite the opposite is the case, that is, the resistivity becomes extremely high at low temperatures, and actually has a minimum as a function of temperature, acquiring a positive temperature coefficient only well above room temperature.

To a first approximation, graphite has been described as a semiconductor with zero energy gap (86), the carriers having very low effective mass in directions in the graphite planes near the points where the bands touch. The fact that the conductivity becomes very high at low temperatures has led to the suggestion that there is a slight overlap of the bands, so that the behavior is metallic (87, 88). For pure graphite crystals of high perfection, the Fermi level is probably midway in the region of overlap of the two bands, so that there are equal numbers of holes and electrons. The electrons have mobility over four times that of the holes, as indicated by a negative Hall coefficient. It is known that the position of the Fermi level can be controlled by the addition of impurities added interstitially in the so-called lamellar compounds (89, 90). Alkali metals, for example, act as donors, raising the Fermi level and increasing the number of electrons, while halogens lower the Fermi level, increasing the number of holes. These atoms go in between the graphite planes. The Hall effect goes through zero for an acceptor concentration of 2.6×10^{-3}, from which the intrinsic electron and hole concentration at room temperature can be deduced as about 6×10^{-4} per atom. Pure graphite has a very high diamagnetic susceptibility which is ascribed to the low effective mass of electrons and holes near the band edges. The susceptibility decreases rapidly with the addition of either donor or acceptor impurities, and since this effect should be independent of scattering, it can be used directly to measure the number of traps in radiation experiments (see below) (91).

Thermal conductivity in graphite is entirely by lattice vibrations, and at low temperatures, arises from transverse vibrations propagated in the planes. Both the specific heat and the thermal conductivity are proportional

to T^3 at low temperatures (92, 93). These results can be explained on the basis of normal mode theory (94). The thermal conductivity result indicates a temperature independent mean free path due to atomic imperfections. In manufactured graphite, the thermal conductivity shows an anomalous temperature dependence which is now ascribed to the effect of the intergranular material (95).

Graphite has rather interesting mechanical properties. The properties of perfect crystals have not been measured, the observed properties being those of manufactured graphite and strongly influenced by microstructure. The modulus is extremely low, comparable to that of lead at room temperature, but increases as much as 25 per cent up to 1600°C., and 100 per cent at 2500°C. Graphite shows some plasticity, even at room temperature, and will support up to 7 per cent elongation at high temperatures (96). As a result, its mechanical strength increases with temperature, up to extremely high temperatures, and it is also ideally resistant to thermal shock, a fact which helps to account for its usefulness as a refractory.

Graphite is subject to oxidation, which takes place at an appreciable rate at 500°C. in pure oxygen, the rate increasing rapidly with decreasing purity. It can also be oxidized at about 700°C. in steam, and 900°C. in CO_2 (97).

Radiation damage, general.—The possible dire consequences of radiation damage to graphite and other reactor materials were first emphasized by Wigner during the early stages of the Atomic Energy program (98). As a result, the problem has come to be referred to in reactor circles as the "Wigner disease." An extensive experimental program, primarily of an engineering nature, was carried on during and after the war on reactor graphite. This program originated in the war-time Metallurgical Laboratory in Chicago, and continued at Argonne, Hanford, Battelle, Knolls Atomic Power Laboratory and North American Aviation. It remained almost entirely classified until the Geneva conference. The results of this classified work have been summarized in the paper prepared for the Geneva conference by Woods and his associates (99). A review of the parallel British work has also been given by Kinchin (100).

Most of the engineering information has been obtained by neutron bombardment in production or other graphite reactors. Large uncertainties exist as to the fast flux which obtained during the exposure, and exposures are usually expressed in terms of an integrated *nvt* which could vary considerably in its damage potentialities. In the discussion which follows, exposures will be expressed in terms of integrated *nvt*, using an equivalence of 1 megawatt day per ton of uranium in a graphite reactor for 5×10^{17} integrated *nvt*. It must therefore be recognized that the actual recoil energy delivered by neutrons to the graphite could have varied by a factor of at least 2 in different cases. The effects on various physical properties are discussed in the following paragraphs.

(*a*) Mechanical properties. As a result of irradiation, the mechanical strength of reactor graphite increases rapidly up to an *nvt* of 10^{20} neutrons/

cm.^{2}, where it is a factor of 3 greater than normal graphite. It gradually falls off with additional exposure. The behavior of Young's modulus parallels this closely, also increasing by a factor up to 3. Exposed graphite becomes very hard, as indicated by its almost total lack of machinability.

(*b*) Thermal conductivity. One of the most dramatic effects of exposure is is on the thermal conductivity, which has been decreased by a factor as large as 50 in some cases. As with many properties, the effect of radiation on thermal conductivity is highly sensitive to the temperature of exposure, as is indicated by Figure 1. The temperature coefficient of thermal conductivity changes sign, so that after exposure, it increases with increasing temperature. One would expect a positive temperature coefficient if the mean free path remained constant, the increase being attributable just to the increase in specific heat. However, the temperature coefficient for the irradiated sample is several times larger than would be predicted from specific heat alone, indicating that the mean free path increases with increasing temperature after bombardment.

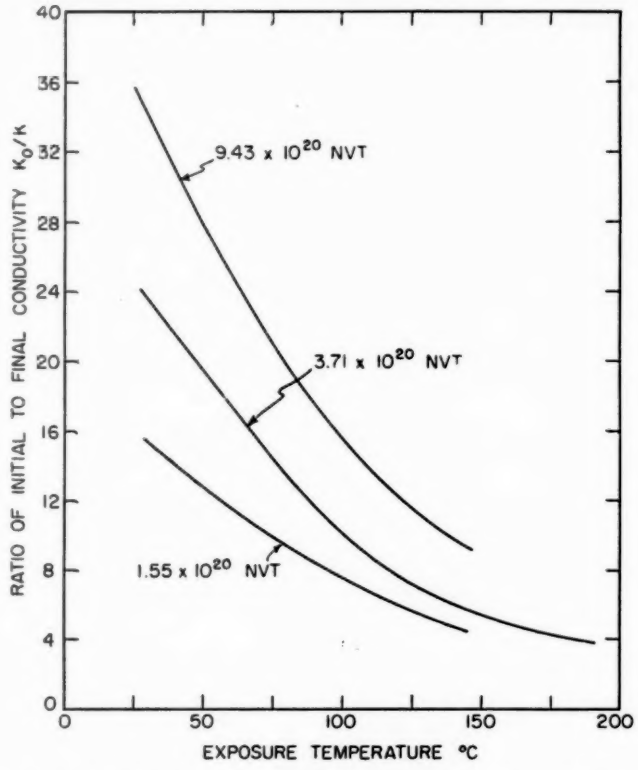


FIG. 1. Effect of exposure temperature on the thermal resistivity increase for neutron bombarded graphite. (From Fig. 6 of Ref. 9.)

(*c*) Electrical resistivity. The behavior of electrical resistivity is in complete contrast to that of thermal resistivity. The change saturates at about a factor of 5 increase and an integrated flux of 2×10^{20} and thereafter stays about constant. In general, electrical resistivity changes are much less sensitive to the temperature of exposure. The difference between electrical and thermal resistivity can be readily explained with the aid of the band model for graphite. The thermal resistivity depends only on the scattering from defects and hence it is increased. The electrical resistivity rises until the number of defects introduced by the radiation becomes about equal to the number of carriers that were present naturally in the original graphite. At this point, each new scattering center introduces a new carrier (that is, a hole) at the same time, and so the effects compensate. The changes in other electric and magnetic properties can also be correlated with this picture, as will be seen later.

(*d*) Crystal structure. Graphite shows rather remarkable changes in crystal structure. These may be described as a pushing apart of the graphite planes, or an expansion along the *c*-axis, accompanied by a slight shrinkage in the planes and probably some warping. The effects are illustrated by the x-ray diffraction results shown in Figure 2. Above an exposure of about 4×10^{20} neutrons/cm.^{2} the x-ray line shape begins to be distorted, and the lines become asymmetrically broadened in the direction of small spacing. At exposures above 10^{21} *nvt* the lines become almost unobservable, indicating that the graphite has reached a nearly amorphous state. The maximum *c*-axis expansion observed is about 16 per cent for 2×10^{21} *nvt* at 30°C. The *c*-axis expansion is nearly as sensitive to exposure temperature as is the thermal

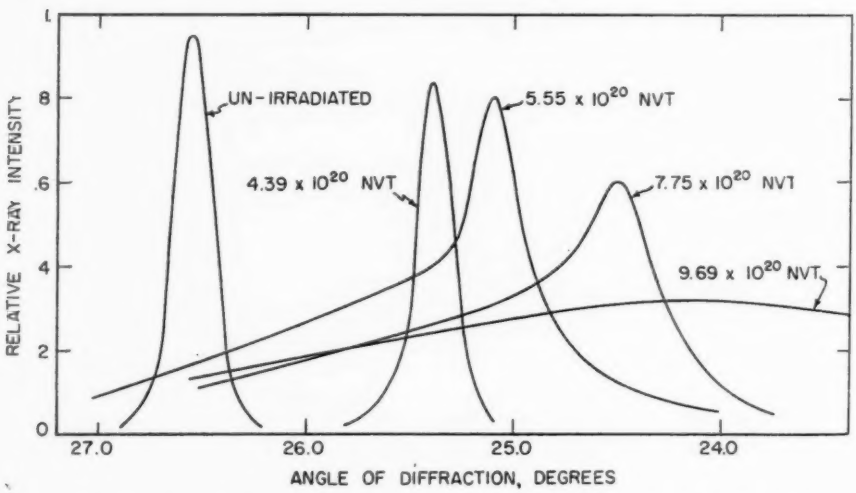
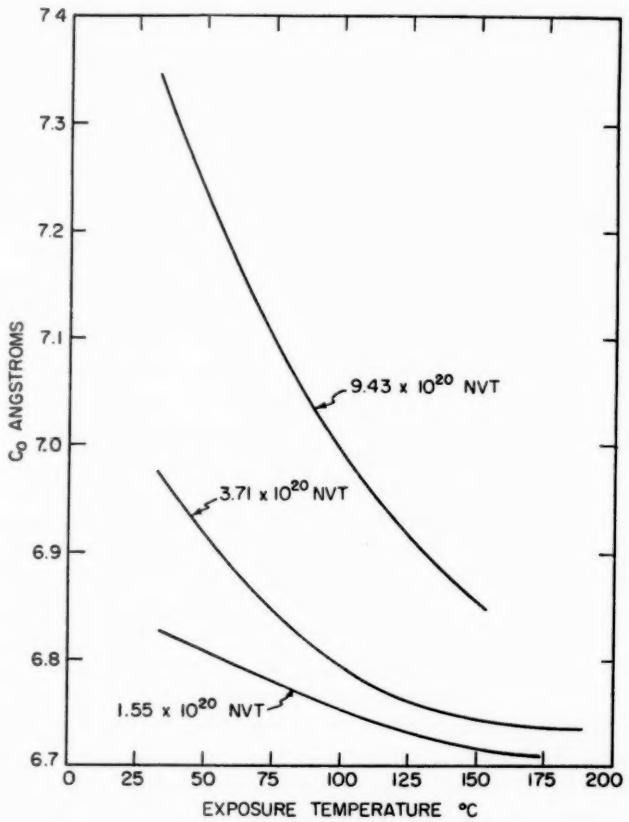


FIG. 2. The (002) X-ray diffraction line for graphite after various amounts of neutron bombardment indicated. (From Fig. 12 of Ref. 9.)

conductivity, and it is the only property which appears to increase linearly with exposure to the highest exposures. A slightly greater tendency towards saturation is indicated, however, at the higher temperatures of exposure. The effect of exposure temperature on expansion is shown in Figure 3.

Keating (101) has investigated the *c*-axis expansion of graphite briquettes, compacted out of natural flake graphite, and irradiated at 80°K. in the Brookhaven reactor. He found a change of 4.75 per per 10^{20} *nvt*, as compared with 2.35 per cent for exposure at +50°C. These values are to be compared with an average figure of 1.7 per cent per 10^{20} *nvt* quoted for reactor graphite, exposed at 20°C. (99). However, the fluxes may not have been comparable.

(*e*) Dimensional stability. One would expect from the *c*-axis expansion that a volume increase would occur, and in view of the anisotropy a change in length in the direction of preferred orientation of the *c*-axis of the crys-



IG. 3. Increase in interplanar spacing of irradiated graphite as a function of exposure temperature for various irradiations. (From Fig. 14 of Ref. 99.)

tallites. Such an effect is observed, but is highly dependent on the manufacturing history, and the preferred orientation of the material. For "transverse cut" graphite, that is material having its long dimension perpendicular to the direction of extrusion, and hence parallel to the *c*-axis, the average expansion rate (99) is about .08 per cent per 10^{20} *nvt* which is about 20 times smaller than the *c*-axis expansion. The linear expansion appears to be proportional to exposure although there is sometimes a break in the curve around 7×10^{20} *nvt*, after which the rate may show either an increase or a decrease depending on the grade of graphite. A transverse cut briquette of compacted natural flake graphite showed an expansion rate of about 1 per cent per 10^{20} *nvt* in contrast with reactor graphite made from petroleum coke. In this case, the physical expansion was close to the x-ray value. Parallel-cut graphite samples having their long dimension in the plane of the crystallite, show an initial expansion on irradiation followed by a contraction. The average volumetric expansion is about twice the linear expansion. The dimensional stability of different grades of graphite shows an excellent correlation with their thermal expansion coefficients. In this connection it is interesting to note that the linear thermal expansion coefficient of transverse cut graphite is about 1/7 of the intrinsic thermal expansion of the crystal in the *c*-direction as determined by x-ray methods (102). This suggests that the relation between the crystal and bulk properties is about the same for thermal expansion and expansion induced by radiation.

Like the thermal conductivity and the x-ray expansion, the dimensional expansion is very sensitive to the temperature of exposure.

(*f*) Stored energy. Together with dimensional stability and thermal conductivity, stored energy is, from the engineering standpoint, one of the most important properties of graphite under irradiation. This energy can be very large, over 6 kcal/mole of graphite for exposures of 2×10^{21} *nvt*. This is about 4 per cent of the heat of sublimation of 170 kcal/mole. The stored energy shows a greater tendency to saturation than the *c*-axis expansion, the rate of increase of stored energy with exposure decreasing to less than $\frac{1}{2}$ of its initial value for exposures above 10^{21} *nvt*. Both these properties are probably closely related to the total interstitial content between the planes. It seems reasonable to suppose that about twice the heat of sublimation is required to place an atom in an interstitial position, since twice as many bonds in the plane are broken. Using this result gives an initial rate of production of interstitials of 0.2 per cent per 10^{20} *nvt*, as compared with 2.4 per cent per 10^{20} computed from the long wave neutron scattering (24). In view of the inaccuracies in the flux, not much significance can be attached to this discrepancy. Also there is evidence of considerable coagulation of defects at room temperature and below.

The nature of the stored energy is most interesting. While the total increases monotonically to the highest exposures, the amount which can be annealed out below 1000°C. actually reaches a maximum of 3.6 kcal/mole at 1.5×10^{21} *nvt* and then begins to decrease. The reason for this is illustrated

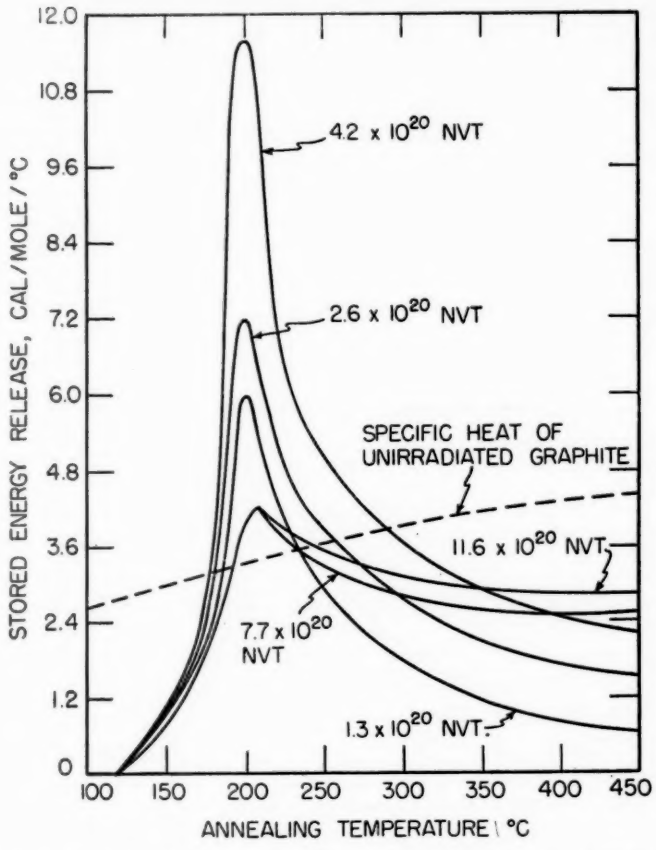


FIG. 4. Stored energy release as a function of temperature for various integrated exposures, as determined by heating at a uniform rate. (From Fig. 30 of Ref. 99.)

in Figure 4, which shows the results of annealing experiments for various exposures. The curves show the stored energy released per degree rise in temperature, for a uniform rate of heating. The prominent annealing peak at 200°C. builds up to a maximum at 4.2×10^{20} *nvt* and then rapidly decreases again for higher exposures. It is interesting that this is exactly the *nvt* for which the asymmetrical x-ray line broadening also begins. After the maximum is reached, most of the damage goes into imperfections which can only be annealed out at very much higher temperatures. This is attributed to coagulation of isolated defects into larger and larger clusters which apparently eventually tend to form new interstitial graphite planes.

The stored energy in the neighborhood of the peak is actually such as to give a negative specific heat to the irradiated material over a small temperature range.

The stored energy annealing characteristics are very sensitive to exposure temperatures, and it is found that for a given temperature of exposure, one has to go to considerably higher annealing temperatures before any stored energy is found, roughly 100°C. above the exposure temperature. This may be regarded as further evidence for the process of radiation annealing (73). Additional evidence is shown quite dramatically by Figure 5. Two conclusions may be drawn from the results shown in this figure. (*a*) A sample exposed at 30°C. and then at 150°C. shows much more expansion than a sample exposed at 150°C. *ab initio*, and (*b*) the sample exposed at 30°C. shows appreciable recovery of damage during exposure at 150°C. A sample similarly exposed at 30°C. must be heated to at least 250°C before any recovery of damage is found in the absence of irradiation (103).

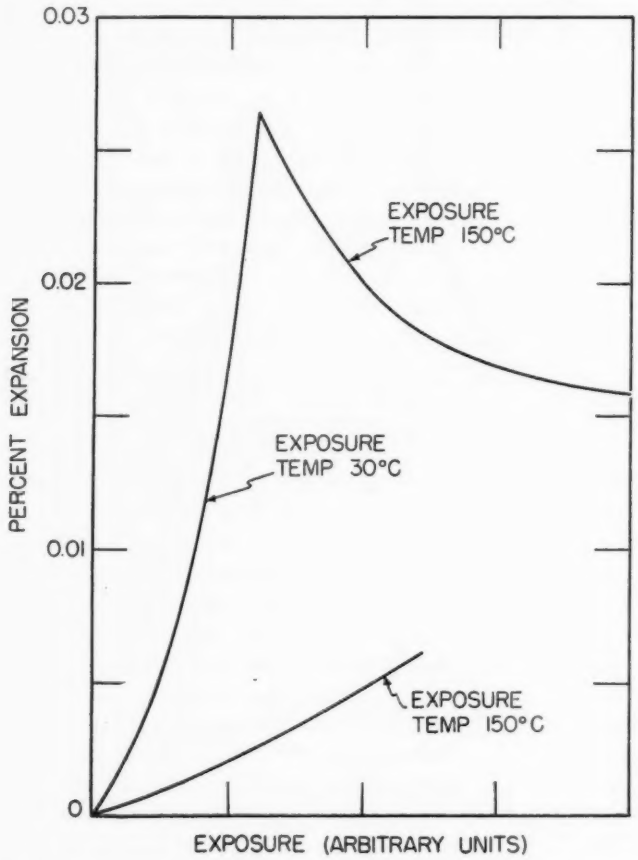


FIG. 5. Radiation annealing of dimensional expansion of graphite. (Taken from Fig. 45 of Ref. 99.)

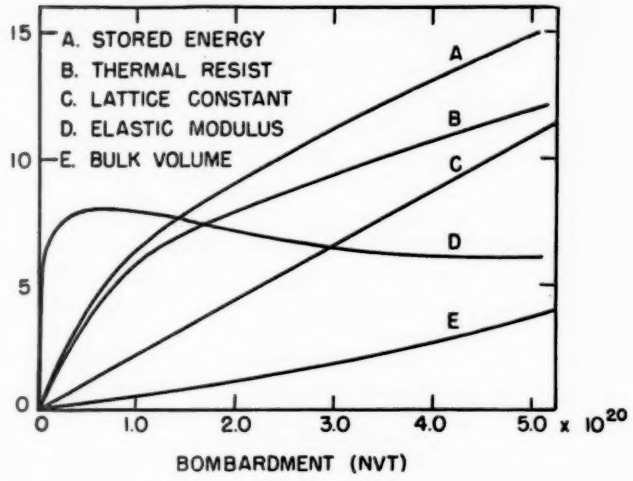


FIG. 6. Summary of changes of various physical properties of graphite with bombardment at room temperature. (From Fig. 3 of Ref. 10.)

The bombardment characteristics of the various physical properties discussed above are summarized in Figure 6.

(*g*) Chemical reactivity. An interesting effect of radiation damage on the chemical reactivity of graphite has been reported by Kosiba & Dienes (104) who have studied the graphite-oxygen reaction between 300° and 400°C. on samples previously exposed to 4×10^{20} neutrons/cm.^{2} at room temperature. This exposure increased the oxidation rate relative to an unirradiated sample by a factor of 6. If the samples were exposed to 2×10^5 rep per hr. of gamma rays during oxidation, the rate was further increased by a factor of 2, presumably due to ionization or dissociation of the oxygen molecules in contact with the graphite. However, little effect of the gamma radiation alone was observed.

Hurst & Wright (105) also report that the rate of oxidation of graphite is accelerated during irradiation in a reactor, The rate is many times the corresponding thermal rate in the temperature range 300° to 350°C. These authors also report a significant acceleration of the rate of reduction of CO_2 by graphite in the range 400° to 450°C.

Tracer experiments described below show that while stored energy increases the chemical reactivity of graphite, the effect is not due to preferential oxidation of the displaced atoms.

(*h*) Other electrical properties. The changes in electrical properties as a function of bombardment are summarized in Figure 7. This figure shows how various electrical properties change with bombardment on a relative scale. Curve A for the electrical resistivity has already been explained in terms of the competition between the scattering due to damage centers and

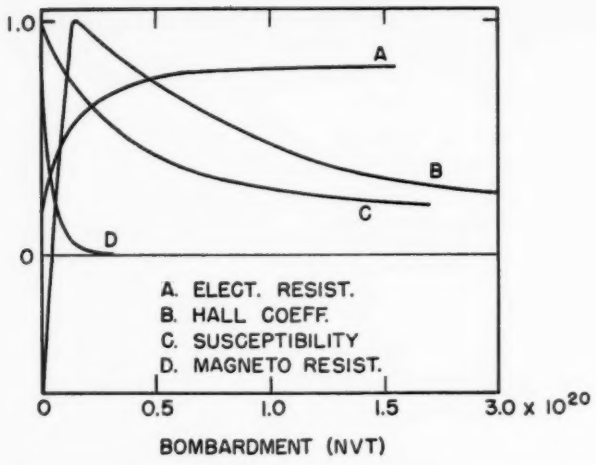


FIG. 7. Summary of changes in electric and magnetic properties of graphite with bombardment (From Fig. 5 of Ref. 10.)

the increase in the number of holes due to electron traps provided by the damage centers. The Hall coefficient can be explained in a similar fashion. As traps are produced, the Hall coefficient goes through zero and reaches a maximum at the point where the number of electrons becomes negligible compared with the number of holes. Thereafter, the Hall coefficient is inversely proportional to the number of holes, and falls off as this increases. The magnetic susceptibility falls off as the number of traps increases, and as has been explained before, can be used directly to compute the number of traps by comparison with its behavior in interstitial compounds (91). The magnetoresistance falls off very quickly as the electron mobility decreases. In the range where only one kind of carrier is present, the product of Hall coefficient and conductivity is a measure of the carrier mobility, and accordingly this is seen to decrease in direct proportion to the number of added traps. At the exposure for which the Hall coefficient vanishes, it can be estimated that 2 per cent electron traps are produced for 10^{20} *nvt*. This is in good agreement with the long wavelength neutron scattering results, and with the *c*-axis expansion if we assume one trap per Frenkel pair.

Paramagnetic resonance.—Bombardment produces traps which exhibit paramagnetic resonance (106). However, the production of such centers saturates almost completely at 3×10^{20} *nvt* for room temperature exposure. Futhermore, the rate of electron trap production as deduced from the diamagnetic susceptibility is at least twice the rate of paramagnetic center production. Paramagnetic centers produced at 80°K. begin to anneal out and hence can migrate at −100°C. They may thus be attributed to interstitials. The mobility of the paramagnetic centers is extremely sensitive to the amount of radiation, indicating that they are very readily trapped at

other defects in the lattice. The resonance shows considerable line broadening at 80°K., and if this is assumed to be due to dipolar interaction, one deduces that the average separation of the centers is only about 10 Å. The line narrows at the temperature at which evidence of migration appears, and this is attributed to diffusion apart of the interstitials to an average separation of about 30 Å. Additional evidence for this dispersal of interstitials comes from the annealing characteristics of the electrical resistivity. This actually shows a slight increase around −100°C., corresponding to the fact that as the interstitials become dispersed, they are more effective as scattering centers.

Low temperature experiments.—Several other properties have been studied with low temperature bombardment, both with protons from the cyclotron at Berkeley, and with neutrons at Brookhaven. Measurements made include stored energy, thermal and electrical resistance, thermoelectric power and magnetic susceptibility. The initial rate of energy storage for all stored energy which can be annealed below 20°C. is about 0.8 kcal/mole for 10^{20} *nvt*. This is the same as the rate of storage of energy above room temperature by room temperature bombardment. If we make the doubtful assumption that the energy storage above room temperature is not changed in the low temperature irradiation, then the total rate of energy storage is doubled for the low temperature bombardment, in agreement with the x-ray results of Keating (101). Annealing of the magnetic susceptibility indicates that only about 20 per cent of the electron traps are removed by annealing up to room temperature. The fact that these show signs of migration at temperatures as low as 100°K. indicates that they have an activation energy for migration of the order of 0.2 ev and are undoubtedly single interstitials.

Tracer experiments.—Henning and coworkers have employed an interesting technique originated by Libby for investigating the fate of displaced atoms in graphite (107). Radiation damage is produced by fast neutrons in a reactor, or by neutrons from the (p, n) reaction on Li in a cyclotron, or by energetic gamma rays from a betatron. In addition to displacements such bombardment generates radioactive C^{11} through an $(n, 2n)$ or (γ, n) reaction on C^{12}. The activated atoms are all displaced because of their high recoil energy, and therefore serve as tracers for all energetic knocked on atoms. The disposition of the C^{11}, and hence of all C interstitials which do not remain close to their parent vacancies, is determined by partial oxidation of the graphite crystals between 400° and 700°C. It can be demonstrated that the oxidation proceeds uniformly inwards from the surface exposed to air, and hence collection and counting of the CO_2 produced gives a method of determining what fraction of the displaced atoms migrate to the exposed surfaces either during bombardment or during any anneal above the combustion temperature. Partial oxidation was also carried out with chrome-sulfuric acid at from 60° to 90°C. The reagent penetrates between the graphite planes and is presumed to oxidize displaced atoms preferentially without regard to their location relative to the surface. It thus provides a method for distinguishing between normal and displaced atoms.

Results of such experiments suggest the following conclusions:

(*a*) Less than 10^{-3} of the displaced atoms migrate to and reintegrate at the exposed surfaces of the crystallites in natural graphite.

(*b*) During annealing above 800°C. about 80 per cent of the interstitials recombine with vacancies and the remainder are trapped at mosaic boundaries and other crystal imperfections.

(*c*) Of the atoms which are displaced far from their vacancies, less, and probably much less, than 8 per cent recombine with vacancies during bombardment or during anneal below 100°C.

(*d*) With increasing annealing temperatures above 400°C. the fraction of interstitials which does *not* recombine falls off rapidly, being about 3.5 per cent at 1000°C., and 0.06 per cent at 1700°C. The last number represents those interstitials which have migrated to the surface, as determined in (*a*).

(*e*) Regardless of the extent of bombardment it can be shown that the residual vacancy concentration above 1000°C. cannot exceed 10^{-6} per atom. The considerable amount of residual damage above this temperature must then be ascribed almost entirely to clusters.

The tracer technique in graphite is valuable primarily because the principal annealing mechanism is migration of interstitials without exchange with the normal atoms. In most crystals, however, interstitial migration takes place by the "interstitialcy" mechanism, in which an interstitial changes places with a normal lattice atom. In such cases the radioactive atom does not act as a tracer for displaced atoms, but becomes rapidly integrated with the normal crystal. This proves to be the case, for example, in diamond (107), for which there was no evidence of long range migration of C^{11} after formation in the lattice.

Discussion.—Hennig & Hove (106) have attempted a comprehensive interpretation of all the graphite experiments along the following lines. Vacancies trap two electrons and are diamagnetic. Two thirds of the interstitials trap one electron and become paramagnetic, the remaining one-third being too close to vacancies to act as traps. Because they both have a negative charge, the interstitials and vacancies find it hard to recombine, and they must surmount an activation barrier. It is this annihilation process which accounts for the prominent annealing peak around 200°C., the annealing below this temperature being largely accounted for by recombination of interstitials into C_2 interstitial pairs through migration of single interstitials. Above about 600°C. all but 20 per cent of the vacancies and interstitials have recombined. The remaining interstitials are trapped at mosaic boundaries, and an equal number of vacancies remain which probably become mobile around 1200°C., in line with the self-diffusion measurements of Kanter (209) and the predictions of Dienes (210).

The evidence is probably not sufficient to support the above explanation uniquely, and it does have some difficulties. For example, it seems unlikely that an interstitial carbon would act as an acceptor, especially when one considers that in valence semiconductors interstitials appear to act as donors.

Furthermore the model implies three traps per Frenkel pair, a value which seems high though not excluded by the present inaccurate data. The resonance data may be equally well explained if the interstitials lost an electron and the resonance were ascribed to an unpaired hole. The only present argument against this is the apparent difficulty of recombination of interstitials and vacancies, which suggests they have like charges. However, the recombination process may be quite complex, so that this argument is not very convincing. The situation is further confused, of course, by the possible existence of aggregates. An experiment which would greatly clarify the situation is a measurement of diamagnetic susceptibility, paramagnetic resonance, and long wave neutron scattering on samples exposed in the same reactor under identical conditions so as to obtain an accurate evaluation of the number of traps and paramagnetic centers per Frenkel pair.

Uranium

Physical properties.—It is, perhaps, indicative of the perversity of nature that two materials of great importance to reactor technology, graphite and uranium, should be unique structurally and highly temperamental in their mechanical properties and radiation behavior. Of the two, uranium is by far the worst. The situation has been aptly summarized by Paine & Kittel in their Geneva paper (108):

> "Candidly, it is not a very satisfactory engineering material for use in a nuclear reactor. It refuses to retain its dimensions and shape during irradiation unless treated with the utmost consideration, and assumes a defiant attitude towards the best efforts to surmount its idiosyncrasies and make it behave like a proper metal."

This perverse behavior is the result of the highly anisotropic crystal structure of the low temperature, or alpha modification, of the metal (109). This phase, which is stable up to 668°C., is based on an orthorhombic unit cell with 4 atoms per cell. The structure may be regarded as composed of corrugated sheets of atoms normal to the (010) direction, with a characteristic nearest-neighbor spacing of 2.8 Å in the (100) direction. The binding between sheets is weaker with a nearest-neighbor distance of 3.3 Å. Uranium in the alpha phase is often described as a semi-metal with a valence of 4, and alloying metals having the same valence, such as tin, zirconium, and hafnium tend to stabilize this phase (110).

There is also a high temperature modification, the gamma phase, stable above 774°C. This is body-centered cubic and characteristically metallic, often described as having a valence of 6 (110). Between the stability ranges of the high and low temperature modifications, there exists an exceedingly complex intermediate structure, known as the beta phase, which is stable only between 668° and 774°C. This has a tetragonal unit cell of 30 atoms (111). The alloying behavior of this phase is suggestive of a valence of 5 (110), and the phase tends to be stabilized by "pentavalent" metals, such as vanadium, niobium, and tantalum. Both the gamma and beta phases may be retained metastably at room temperature by quenching suitable alloys,

for example, alloys containing about ten atomic per cent zirconium or molybdenum. The beta phase can be retained in low chromium alloys, and with still lower solute concentrations a martensite-like transformation from beta to alpha has been observed (112).

Pure uranium, as used in reactors, is always in the alpha phase. The anisotropy of this structure is especially evident in its thermal expansion properties. The thermal expansion is actually negative in the (100) direction which corresponds to the longest dimension of the unit cell. Pure alpha uranium is quite soft, especially at elevated temperatures, and it recrystallizes above 400°C. for moderate deformations (109). The strength of this phase can be markedly increased by alloying additions (110).

Deformation of uranium occurs by both slip and twinning. The primary slip system is (010)-[100] with a critical shear stress of 0.34 kg./mm.^{2} determined on single crystals. Twinning on a (130) composition plane also occurs and represents an alternate deformation mode especially at lower temperatures (109, 113). The yield stress decreases rapidly with increasing temperature. Cold work produces a preferred orientation in which the (010) axis tends to line up along the working direction, i.e., the rolling direction or the extrusion axis. As the temperature of deformation is increased, a (110) texture tends to predominate. The degree of (010) orientation is roughly indicated by the smallness of the thermal expansion in the direction of work but in no case is the texture perfect enough to give a negative expansion coefficient.

A few of the important physical properties of uranium are listed in Table I (109).

TABLE I

PHYSICAL PROPERTIES OF URANIUM

X-ray density (25°C.)	19.04 gm./cm.^{3}
Transformation temperature	668°C.
Transformation temperature	774°C.
Melting point	1132°C.
Cohesive energy at 0°K.	116.6 kcal./mol.
Thermal conductivity (70°C.)	0.297 watts/cm./°C.
Specific heat (25°C.)	6.612 cal./mol./°C.
Thermal expansion	(100) 36.7×10^{-6}/°C.
	(010) -9.3×10^{-6}/°C.
	(001) 34.2×10^{-6}/°C.

Thermal cycling.—One of the most striking properties of alpha uranium is its remarkable dimensional instability under both thermal cycling and irradiation. This instability manifests itself in elongation in the direction of prior working, in surface roughening and in the development of subgrains and occasionally of porosity (114). The irradiation and thermal cycling behavior are similar in many respects, but cannot be identical as indi-

cated by the fact that single crystals grow under irradiation but are stable under thermal cycling (108).

The elongation is the most dramatic property, and appears to continue almost idenfinitely with cycling. For example, six fold increases in length have been produced in alpha-rolled uranium, subject to 3000 cycles between 50 and 550°C.

Growth rates are often shown graphically by plotting the natural logarithm of the growth ratio L_N/L_0 as a function of N, the number of cycles. Here L_N is the length after N cycles and L_0 the initial length. The tendency to growth is expressed in terms of the growth coefficient G_t, which is the slope of the $\ln(L_N/L_0)$ vs. N plot. It is found in practice that the growth follows a law of the form:

$$\ln \frac{L_N}{L_0} = aN + bN^2 \qquad 16.$$

so that the growth coefficient increases linearly with the number of cycles, at least up to 800 cycles. At much higher values of N, G_t begins to decrease again.

The growth rate G_t is influenced by both metallurgical and cycling variables, which are listed briefly below (114).

(*a*) Preferred orientation. The growth rate increases with preferred orientation, and is favored by (010) texture. Thus alpha rolled rods of uranium show much greater growth if the deformation is carried out at room temperature than if carried out at 600°C. The slower growth for the higher temperature deformation reflects the tendency towards (110) texture under these conditions. Whereas rolled material shows a G_t of 500 to 600, material heated into the beta range and then cooled shows a G_t of only 20 to 30. Since the degree of preferred orientation increases with percentage deformation, the growth coefficient does also, as indicated in Figure 8b.

(*b*) Grain size. Fine grained material has a much higher growth coefficient than coarse grained material, as indicated by Figure 8a, and the sensitivity to grain size increases with the degree of preferred orientation. The acceleration of the growth rate as a function of the number of cycles is attributed to a break up of the alpha grains into sub-grains which occurs as a result of thermal cycling. Coarse-grained material shows a much greater acceleration than fine grained material. Texture and grain size also affect the growth under irradiation, as indicated by Figure 8b, but the sensitivity to grain size is much less in the case of radiation.

(*c*) Chemical composition. Low concentrations of alloying elements have little influence on reducing the growth rate of uranium worked in the alpha phase. In some cases, they may even increase the growth rate indirectly through producing grain refinement. In other cases they contribute to randomizing the orientation and reduce growth. If the composition is such that the material is retained in the beta or gamma phase, thermal cycling is prevented.

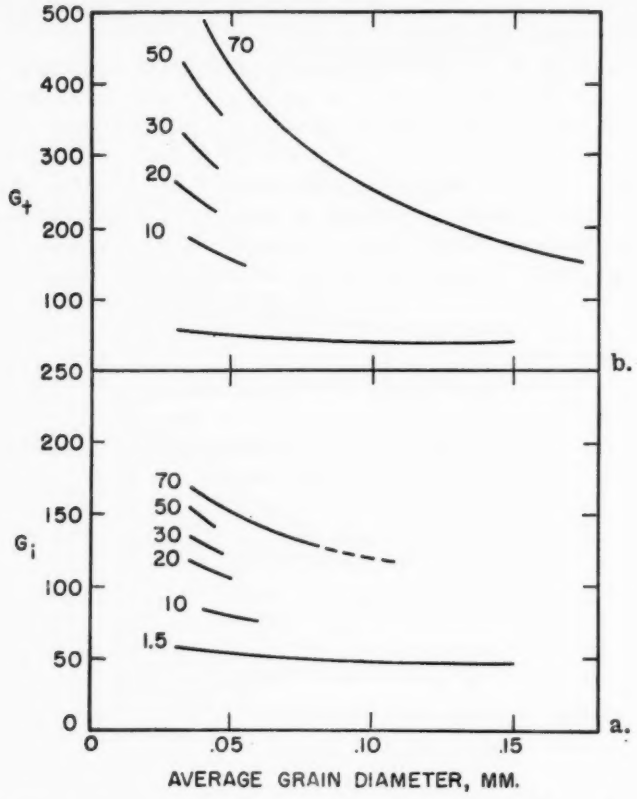


FIG. 8. Effect of grain size and prior cold work on the thermal cycling and radiation growth of alpha uranium. (a) Growth coefficient for thermal cycling. The number on each curve refers to percent reduction by rolling prior to final anneal to recrystallize in alpha range. (b) Growth coefficient for irradiation. Deformation and recrystallization treatments were the same as in (a). (From Figs. 2 and 3 of Ref. 10.)

(*d*) Cycling variables. The greatest growth rates are observed when slow heating and fast cooling are combined with a large temperature range of cycling. In general, the growth rate is much more sensitive to what happens in the high temperature range, and appreciable growth can be obtained even by exposing highly oriented uranium to many small thermal fluctuations in the high alpha temperature range.

Thermal cycling also produces other effects, all of which are indicative of the large stresses to which the grains are evidently subjected as a result of their effort at mutual accommodation. These effects include polygonization, crystallographic slip, especially near boundaries, roughening of the boundaries and boundary migration. In addition to this, the development of poros-

ity accompanied by a decrease in density is observed, especially in less pure material. However, the porosity does not appear to be fundamental to the growth mechanism, as in pure materials growth occurs without it. Slip is the most pronounced in coarse grained specimens and twinning also occurs. Grain boundary migration is most evident in fine-grained specimens. Cycling produces no detectable change in preferred orientation, and there is no evidence for growth of individual grains in a preferred direction.

No entirely satisfactory explanation of the thermal cycling growth has been put forward. Suggestions are of two types. One mechanism involves nonreversible plastic flow under the influence of the highly anisotropic thermal expansion. One such mechanism, termed "thermal ratcheting" has been proposed by Burke, Howe, & Lacey (115). The common idea of all these models is that stress relaxation occurs by a different mechanism at high and low temperatures. Burke *et al.* suggest that deformation is by grain boundary slip at high temperatures, and by crystallographic slip at low temperatures. The objection to this mechanism is that it predicts elongation of the grains, which does not appear to take place in practice. The other suggested mechanism, that of Fisher (116), involves temperature dependent anisotropic diffusion which results in pumping vacancies from one grain boundary to another. This model predicts a change in grain shape and in addition would predict thermal cycling growth of single crystals, which is not observed.

Irradiation effects.—At Geneva, American work on radiation effects in uranium was reviewed by Paine & Kittel (108), British work by Pugh (117), and Soviet work by Konobeevsky *et al.* (118). These reports constitute almost the only references in the open literature.

In analogy with thermal cycling growth, one can define a radiation growth coefficient G_i, which is equal to:

$$G_i = \frac{\ln (L_t/L_0)}{\text{fraction of atoms fissioned}} \qquad 17.$$

Single crystals of alpha uranium show substantial growth under irradiation, but no growth with thermal cycling. They elongate in the (010) direction, and shorten about equally in the (100) direction, remaining unchanged in the (001) direction. The magnitudes are shown in Table II. The growth observed

TABLE II

RADIATION GROWTH COEFFICIENTS FOR ALPHA URANIUM SINGLE CRYSTALS AT ROOM TEMPERATURE

Direction	G_i
(100)	-420 ± 20
(010)	$+420 \pm 20$
(001)	0 ± 20

in imperfect lineage crystals made by gradient transformation from the beta to the alpha phase, is even greater (108). Polycrystalline materials show growth to the extent that the (010) preferred orientation is developed by prior fabrication. The effects of grain size and percentage of prior deformation are indicated in Figure 8b. All these results are for specimens rolled and subsequently recrystallized at 575°C. Materials rolled at 300°C. and irradiated as rolled show nearly two and a half times the growth rate of material which is rolled and recrystallized before irradiation. The fact that the rate of growth for polycrystalline material can be substantially greater than for single crystals shows that grain boundary restraints do play a significant role but not the exclusive one. Also, it is evident that irradiation growth is much less sensitive to grain size, although the sensitivites to prior work are comparable. As with thermal cycling, rolling above the recrystallization threshold of 400°C. develops a strong (110) component in the texture and thus results in considerably reduced radiation growth. Heat treatment of alpha rolled material into the beta or gamma phase randomizes the orientation and reduces the growth rate to $G_i = 15–20$.

Composition has a considerable influence on radiation growth. In high uranium alloys, this results from two effects. On the one hand, alloying addition can be used to promote more random orientation, as in the case of 0.1 to 0.2 per cent chromium. On the other hand, larger alloying additions may be used to retain the gamma phase at room temperatures. This can be done particularly with molybdenum, niobium, and zirconium and 10 per cent molybdenum in uranium is reported to give excellent dimensional stability. Furthermore, the Russian workers (118) report that irradiation actually promotes randomness in a uranium 9 per cent molybdenum alloy, so that even the two phase alloy is homogenized by radiation. This means that such an alloy should have excellent long term stability.

The definition of a growth coefficient implies that the growth is an exponential function of burn-up, which is indeed found to be the case, to a good approximation.

Pugh has discussed the effect of temperature of irradiation on growth (117). The growth rate apparently attains a maximum between 200° and 350°C. and decreases at both higher and lower temperatures, disappearing almost entirely above 500°C. Kunz & Holden (108) have shown that growth is greatly reduced when the sample is irradiated at liquid nitrogen temperatures.

Growth under irradiation is by far the most dramatic effect. Other physical property changes are observed, however. Surface distortions result from irradiation and are strongly dependent on grain structure. For example large grain, random material such as cast uranium, develops blistering which is so severe that the contours of the specimen are rapidly destroyed. Fine grain material roughens, but retains its shape. Alpha worked material develops longitudinal striations and ridges resembling the grain of wood. In spite of the large dimensional changes, the net volume change is very small, being

about 2 per cent for 1 per cent burn-up. Only half of this change, however, can be accounted for by accumulation of fission products (108, 117). Only a slight increase in x-ray lattice parameter has been observed (108).

The distortion of uranium under irradiation was particularly strikingly revealed in an experiment reported by the Soviet workers (118). They irradiated a sheet of cold rolled and annealed uranium sandwiched between magnesium and stainless steel. After the irradiation, the boundary of the sheet adjacent to the magnesium became undulated, so that the sheet thickness varied from .07 to .45 mm., as compared with an initial thickness of about 0.3 mm. This is probably an extreme example of the graininess produced in alpha worked material, and was further enhanced by the restraints against expansion. The wrinkling phenomenon has also been emphasized by the British workers (117).

Mechanical properties change drastically after irradiation. Burn-up to the extent of .035 atom per cent at 100°C. for example, decreases the ultimate stength by 27 per cent, raises the yield strength by 120 per cent and destroys the ductility (108). The ductility change cannot be restored by annealing in the alpha recrystallization range which is taken as evidence that the mechanical effects result principally from accumulated fission products in the lattice rather than from displaced atoms. Indeed, annealing appears to decrease the ultimate tensile strength, probably owing to the diffusion of fission product gases to microscopic cracks in the material. The Soviet workers (118) have reported about a fourfold decrease in impact strength as a result of irradiation.

Thermal conductivity is decreased both during and after irradiation, as indicated, respectively, by measurements by Woods and Jones and by Siegel & Billington (108). A small increase in the electrical resistivity is observed (108, 118). X-ray line broadening has been reported by Tucker & Senio and is attributed to strains introduced by the fission products in the lattice (119). On the other hand, the Soviet workers (118) have reported an opposite effect in a rolled uranium foil. In this case, they attribute a line narrowing to radiation annealing of the internal strains produced by the prior rolling.

Interesting ordinary and electron micrographs or irradiated uranium have been obtained by the Soviet workers (118) using replica techniques. This work gives evidence of a large amount of twinning within the grains and also reveals a peculiar globular structure with dimensions on a micron scale.

Little progress has been made towards developing a quantitative mechanism to explain the effects of irradiation in uranium. Pugh (117) has given the most extensive discussion of this subject in the open literature. He originally suggested a mechanism involving local plastic yielding by twinning in the vicinity of a fission spike. The important hypothesis is that a different mode of deformation is operating on the expansion and contraction phases of the short heating cycle. Since slip is geometrically reversible, it alone cannot account for continuous growth, and Pugh suggests that the disappearance

of radiation growth above about 350°C. occurs because the yield strength of the material drops to the point where slip occurs in preference to twinning in both phases of the thermal spike. The decrease in growth below 200°C. is attributed to increased yield strength and consequently less yield per spike. The main difficulty with the twinning mechanism is connected with the time available for flow in the short duration of the thermal spike, and lack of detailed understanding of the mechanism of initiation of the twinning process in sub-microscopic dimensions.

Seigle & Opinski have suggested an alternative mechanism based upon anisotropic diffusion of displaced atoms and vacancies, created by fission recoils (108, 116). It is postulated that interstitials form new close packed rows parallel to the (100) direction between the corrugated sheets. This forces the sheets apart, and thus accounts for expansion in the (010) direction, which amounts to a mechanism similar to that suggested for graphite expansion. At the same time, vacancies in the close-packed (100) rows diffuse along the rows until they reach a grain boundary or dislocation and thus produce shrinkage along this direction. The growth rates with this mechanism might be expected to decrease either when the temperature was so high that vacancies and interstitials mutually annihilate as fast as they are formed, or when the temperature is so low that significant migration cannot occur. For the diffusion mechanism it can be calculated that 100 per cent growth will occur when 50 per cent of the atoms have taken up interstitial positions, and an equal number of vacancies have diffused out of the (100) row. Less than 1 per cent of the vacancies and interstitials produced by fission need to be used in order to produce the maximum observed growth. The chief difficulty with mechanisms of this sort is that the extent of damage is not a simple linear function of the integrated *nvt*, since competititon between first and second order processes is involved. Considerable light on the mechanism of radiation damage in uranium might be thrown by light charged particle studies on natural uranium at low temperatures. This would help in the identification of the various types of defect produced by irradiation and in the determination of the relative mobility of vacancies and interstitials. Until such studies are made, speculations concerning the mechanism of growth are very difficult either to prove or refute.

Irradiation of alloys.—The effects of irradiation on uranium rich alloys have already been discussed. A very fruitful approach to the reduction of radiation damage in fuel is to alloy the uranium in a dilute solid solution in another metal whose structure is such that it is much more resistant to radiation damage than pure uranium. One of the most useful of such alloys has been that of uranium and aluminum. Up to 30 per cent by weight uranium, the microstructure of these alloys is such that the aluminum may be considered as furnishing a continuous matrix with the uranium uniformly dispersed in it. Up to nearly 1 per cent of total atoms fissioned, the changes in properties produced are relatively innocuous (119). For example, dimensional changes are less than 1 per cent, thermal resistivity in-

creases by about 60 per cent, electrical resistivity by about 100 per cent, and there is an increase in hardness. In the 15 per cent by weight alloy six-fold increases in yield strength and ultimate tensile strength have been observed. On the other hand, ductility is drastically reduced, but not destroyed entirely. Similarly good properties are found for uranium-zirconium and uranium-thorium alloys. The interesting thing to note about all these results is that hardly any damage occurs after 1 atom in 10^3 has fissioned, while in pure uranium, drastic effects, for example 5 to 10 per cent elongation are observed when only one atom in 10^4 has fissioned.

Ceramic fuels.—Mixtures of uranium oxide and graphite were extensively studied by Hunter at Oak Ridge (119). Their sensitivity to irradiation proved to be highly dependent on particle size, and to obtain a useful material it was necessary to keep the size of the U_3O_8 particles comparable with the fission product range, so that recoil damage was confined to the particles themselves. Fission damage in the graphite was found to be inversely proportional to particle diameter for small particles. Changes in properties resembled those observed with fast neutrons in graphite, except that coalescence of defects occurred at a very early stage, making annealing difficult.

Other fuel element designs which employ the principle of dispersing fissionable material in a matrix having high resistance to damage have been discussed by Weber & Hirsch (120).

DAMAGE TO VARIOUS SOLID TYPES

Valence Semiconductors

Extensive work has been reported on valence semiconductors during the past four years. The most detailed quantitative work has been on germanium, with somewhat less extensive work on silicon and intermetallic compounds of the III-V type. Work with fast neutrons has been carried on largely by the group at Oak Ridge. Work with deuterons, alpha particles and electrons has been conducted by the group at Purdue, while electron bombardment work has also been reported by a group at Bell Laboratories, and from RCA working with a Van de Graaf machine at MIT.

The model of James and Lark-Horovitz.—In order to set the semiconductor bombardment work into a theoretical framework, it has been convenient to use a picture proposed by James and Lark-Horovitz in 1951 (36). The form which this model has assumed as a result of recent experimental work is shown in Figure 9, taken from a recent paper by the Oak Ridge group (45). According to this model, the damage consists largely of separated Frenkel pairs. The interstitials act as double donors giving rise to levels A and B of Figure 9, which correspond to single and double ionization, respectively, of the interstitials. Similarly, the vacancy is capable of acting as a double acceptor, giving rise to levels C and D in the diagram, which may be regarded as the second and first ionization levels for holes into the valence band. The positions of the levels are those appropriate to germanium and are shown

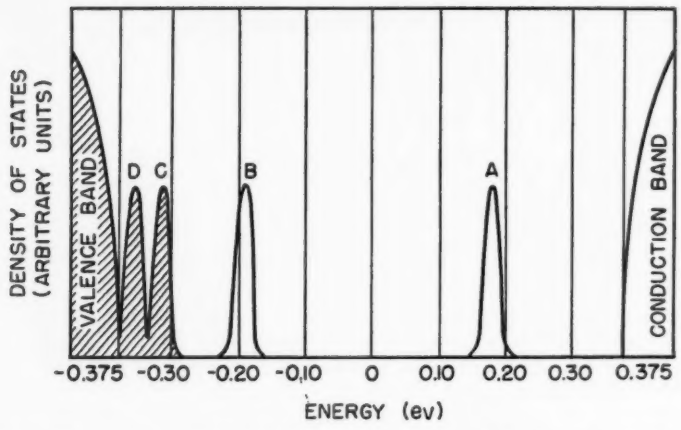


FIG. 9. Proposed energy level diagram for bombarded germanium. (From Fig. 12 of Ref. 45.)

with a hypothetical spread of energies which results from the interaction of the defects. Thus a displacement gives rise to a total of 4 levels in the forbidden gap, although in general only two at a time will be important.

In *n*-type material, the two electrons from an interstitial will fall into the two acceptor levels of a vacancy. The empty interstitial levels will result in two electron traps for each Frenkel pair created. The deepest interstitial level will always remove one electron from the conduction band, while the higher interstitial level, about 0.2 ev below the conduction band, will be only partially effective as a trap, depending on the temperature.

In *p*-type germanium, the situation will be rather different, for the levels are crowded towards the valence band. For *p*-type material it is most convenient to discuss the behavior in terms of hole trapping, with A and B regarded as acceptors of holes, and C and D as donors. However, B is a much shallower hole trap for *p*-type material than C is an electron trap for *n*-type material. Thus, while A and D may be regarded as definitely compensating each other, B and C will contend for mastery of the situation in such a way that bombardment can either increase or decrease the number of holes, depending on the initial hole concentration and the temperature, with the result that bombardment will decrease the hole concentration if it is initially high enough, and increase it if it is low enough.

This general picture is in good agreement with the results of neutron bombardment experiments on both *n* and *p*-type germanium of widely varying initial carrier concentrations (35, 45). All the experiments are carried on in a temperature range such that ordinary donors and acceptors are completely ionized, and the equilibrium concentration of minority carriers can be neglected.

For *n*-type material, the bombardment removes between 1 and 2 carriers

per defect, lowering the Fermi level until the conductivity reaches a minimum at approximately the intrinsic condition. At the minimum, the number of Frenkel defects introduced must be equal to the initial electron concentration. This follows from the fact that all except the highest trap is below the intrinsic Fermi level position. The initial removal rate in terms of electrons per defect can then be calculated on the assumption that the damage varies linearly with *nvt*. Over a wide variety of initial concentrations, the removal is consistent with the assumption that the highest trap lies 0.2 ev below the conduction band. The same level is also readily identified in electron bombardment (122, 123) and with deuteron bombardment (40) and its position can be confirmed by measurements of Hall effect or resistivity as a function of temperature below room temperature after bombardment (35).

In *p*-type material, room temperature neutron bombardment results in a competition between the two shallow acceptor levels, located at 0.18 and 0.066 ev above the valence band, respectively. The rate of hole removal is exactly zero if the Fermi level lies midway between these two levels, since one is a donor and the other an acceptor. Thus the positions of the levels can be accurately deduced from studies of the removal rate as a function of initial carrier concentration.

For *p*-type material, there appears to be a difference in the position of the levels resulting from neutron bombardment on the one hand (45), and electron bombardment on the other (15, 123), with deuteron bombardment giving intermediate results. In the case of electron bombardment, the deeper hole trap appears to lie at 0.26 ev from the valence band, and for deuteron bombardment, at about 0.22 ev (124).

Prolonged bombardment of either *n* or *p*-type Ge ultimately produces material which is *p*-type with a definite carrier concentration such that the Fermi level lies at about 0.123 ev above the valence band, and with an

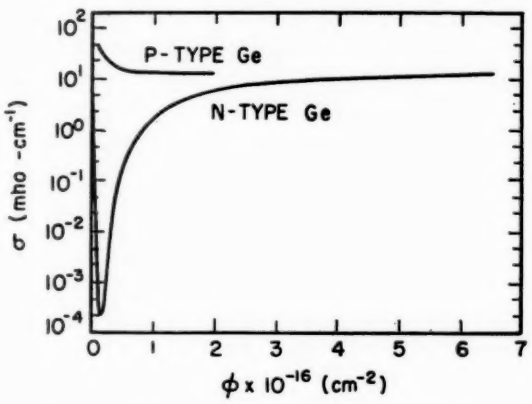


FIG. 10. Effect of prolonged deuteron bombardment on the conductivity of *n* and *p*-type germanium. (From Fig. 7, page 317 of Ref. 124.)

asymptotic resistivity of about 0.1 ohm cm. (124, 125). This is shown in Figure 10. Similar effects occur in silicon, except that the asymptotic resistivity at room temperature is about 10^6 ohm cm., corresponding to an ultimate Fermi level very close to the center of the forbidden gap, about 0.07 ev below the intrinsic level (126). This in turn means that the levels A, B, C, and D for Si are much more symmetrically distributed in the gap than for Ge, a result which is consistent with the fact that chemical impurities such as Au tend to produce levels closer to the conduction band in Si than in Ge (127).

The initial rate of carrier removal has been used to study the level structure in neutron bombarded InSb (128) and GaSb (129). Studies of Hall coefficient as a function of temperature have also been carried out on irradiated samples. In InSb the effects are complicated by the high neutron capture cross section of In, which results in transmutation to Sn. Irradiation in a purely thermal flux showed that the change in carrier concentration was that to be expected if the transmuted Sn occupied a normal In site and acted as a chemical donor impurity (130). On the other hand, samples shielded with In and Cd to minimize transmutation showed conversion from p to n-type, suggesting the production of shallow electron traps in n-type material with the center of gravity of the donor and acceptor traps lying above the center of the forbidden gap (131). In this case the level structure was attributed to vacancies and interstitials in the same way as for Ge and Si, with four types of donor interstitial levels and four types of acceptor vacancy levels corresponding to the two atomic species in the crystal. There is also evidence of shallow hole traps. For GaSb the defect level structure appears to be very similar to that of Ge, as one might expect from the close similarity of other properties (130). The initial carrier removal rate is much more sensitive to carrier concentration in the p-type range than in the n-type range as might be expected if the deeper of the two interstitial donor levels were below the center of the gap.

Carrier mobility.—The interpretation of the initial rate of carrier removal by bombardment is based on the assumption that initially the contribution of defect scattering to the total scattering probability can be neglected. This is a valid assumption for relatively high temperatures and not too high initial carrier densities. However, if the scattering in the semiconductor is already due to ionized impurities before bombardment, then scattering as well as carrier removal will contribute to the change in resistivity. If the level picture of Figure 9 is assumed, then for each Frenkel pair we will have one doubly charged scattering center and two acceptor states. If the scattering is already entirely due to impurities, the apparent rate of carrier removal will be three times the value observed when the scattering is entirely due to lattice vibrations. For both deuteron (40) and neutron (35) bombardment, it is found that the resistivity change at 90°K. is equivalent to the removal of about 5 carriers per Frenkel pair, and is thus about $2\frac{1}{2}$ times what one would expect on the basis of the rate of production of traps

as deduced from the bombardment necessary for conversion from *n* to *p*-type. This is reasonably consistent with the idea that a large part of the resistivity change at liquid N_2 temperature is due to scattering by doubly occupied vacancies.

However, a more detailed examination of the mobility changes indicates that the picture, especially for neutrons, is not very simple. Some of the facts which do not fit the idea of simple ionized impurity scattering are the following:

(*a*) At dry ice temperature, a much greater neutron exposure is needed to produce a changeover from lattice to impurity scattering mobility for high initial carrier concentrations than for low. This suggests that the defects are much less effective as scatterers when a high concentration of carriers is present. Shielding of the electrostatic fields of defects by carriers is to be expected even for isolated scatterers, but the observed effect is much bigger than predicted and suggests that appreciable aggregation of defects takes place at high bombardment (35).

(*b*) At liquid N_2 temperature, the Hall mobility decreases more rapidly than linearly with the integrated flux for *n*-type Ge (132), and the decrease is faster the lower the initial electron concentration. On the other hand, for *p*-type Ge the reciprocal mobility at 77° K. appears to be quite accurately proportional to the defect concentration (133).

(*c*) For ionized impurity scattering one would expect the maximum mobility to be shifted towards higher temperatures for a higher concentration of defects. In *p*-type Ge, this is not observed, but on the contrary, the contribution of the defects to the scattering probability seems to be nearly twice as great at room temperature as at liquid air temperature, and the maximum is not shifted with temperature (134). For *n*-type Ge, the shift in the maximum is towards higher temperature with increased bombardment as expected (135).

(*d*) For *n*-type Ge the electron mobility of bombarded material increases with temperature much more rapidly than the theoretical $T^{3/2}$, and the temperature variation is a strong function of the carrier concentration (136) and of bombardment.

For charged particle bombardment, the behavior of the mobility is more in conformity with theoretical expectation, although the situation has not been analyzed in as complete detail (137). Cleland *et al.* (35) have attributed these differences to the greater clustering of defects which is expected to occur in the case of neutron bombardment. The sensitivity of the scattering to electron concentration and the very strong and erratic temperature dependence of the mobility both point in this direction. On the other hand, such a picture points to no obvious explanation of the difference between hole and electron mobility.

Annealing of damage.—Although the annealing of radiation damage has been extensively studied for Ge and Si, a clear picture of the various processes has not yet emerged. In the case of electron bombardment, the damage

probably consists mostly of simple Frenkel pairs, since the electron does not have energy to produce more. Brown & coworkers (138, 122) have made a serious attempt to analyze quantitatively the annealing of damage produced by 3 Mev electron bombardment of Ge at room temperature. These authors considered three annealing processes (138):

(*a*) short range recombination of vacancy-interstitial pairs,

(*b*) random walk, presumably of an interstitial, followed by recombination with a different vacancy from the one at which it originated, and

(*c*) capture of point defects by surfaces or dislocations.

Even though a single elementary activation step, the jump of an interstitial, is involved, the theory shows that because (*a*) is a monomolecular process while (*b*) is bimolecular, the annealing process should be very extended in time. In agreement with this idea, Brown *et al.* (122) found experimentally that the ratio of times for 90 per cent and 10 per cent recovery was about 10^5. In spite of this, the isothermal recovery curves for five different temperatures could be superposed by an adjustment of time scale following a simple activation law with an activation energy of 1.6 to 1.8 ev, depending on temperature. This activation energy was identified with a single interstitial jump. The best fit was obtained by taking into account processes (*a*) and (*b*) only, as this gave the most reasonable frequency factor for the elementary step. The slight variation of the activation energy between 90°K. and 300°K. could be attributed to the reduction of activation energy for very close pairs. The same authors also studied the damage produced at 78°K. (139). They found evidence for minority carrier trapping (140) in both *n* and *p*-type Ge, and noticed that electron traps annealed at 185°K., while the hole traps annealed at 140°K. There is ample evidence of low temperature annealing from other work. For example, Cleland *et al.* (141) have found evidence for annealing of neutron damage in *p*-type material at temperatures as low as 120°K. Furthermore, the changes in conductivity associated with this annealing are apparently also associated with minority carrier traps. The low temperature annealing process in *p*-type material results in further reduction in conductivity, and is attributed by Cleland *et al.* (141) to coagulation of interstitials. They suggest that the interstitials are actually stabilized by capturing electrons, and that the rate determining step in the coagulation process is the thermal release of a trapped electron. Similar effects are observed in *n*-type material, but at somewhat higher temperatures (35). The low temperature process is sharply distinguished from irreversible increases in conductivity which appear to occur at a maximum rate at around 210°K. in *p*-type material. It is evident from these examples that the recovery effects are exceedingly complex, and much more careful study, especially of electron bombarded material, will be required to untangle the various steps and identify the defects. The low temperature results are not necessarily in contradiction with the theory of Fletcher & Brown (138) since these authors did not include combination of interstitials into pairs as a possible process. This would be especially im-

portant in neutron bombarded material where the defects tend to be densely clustered.

In connection with the study of bombardment produced defects, it is interesting to note that Schulz-Dubois *et al.* at Purdue (142) have recently reported the observation of a paramagnetic resonance line identified with a bombardment produced defect in Si. This line narrows and weakens with partial annealing in a manner analogous to the resonance in graphite. This may prove a very fruitful tool for the identification of defects in Si; unfortunately, the spin resonance cannot be observed in Ge because of its higher spin-orbit coupling (143).

Photoconductivity.—Stoeckman and coworkers at Purdue (144) have made a study of photoconduction in Ge bombarded with 6 Mev electrons at 90°K. In *n*-type material they have found photoconduction bands with thresholds at 0.35 ev and 0.55 ev. These are presumably associated with photoexcitation of electrons from levels A and B of Figure 9 to the conduction band. If the bombardment is sufficient to convert the material to *p*-type, the 0.35 ev band disappears, because this level is no longer occupied. Furthermore, illumination in the 0.55 ev band now quenches the photoconductivity. This result is striking confirmation of the level picture of James and Lark-Horovitz, since level B belongs to an interstitial and therefore acts as a minority carrier trap in *p*-type material. Illumination in this band thus suppresses the trapping and hence quenches the photoconductivity, as observed.

Metals

Mechanical properties.—The effects of radiation on the mechanical properties of metals have recently been reviewed by Dienes (2) and by Billington (119). These effects may be very briefly summarized as follows: (a) The hardness and critical shear stress of all metals is increased; (b) there is a significant decrease in ductility but usually with an increase in the ultimate tensile strength; and (c) there are small increases in the elastic constants.

The critical shear stress appears to be a quite sensitive property for well annealed materials. Blewitt & Coltman (145) have emphasized the analogy between radiation hardening and the hardening in solid solution. Blewitt estimates, however, that one Frenkel defect is about 40 times as effective as a Zn atom in hardening Cu. This is reasonably consistent with the much greater effective strain produced by an interstitial in the lattice, which is equivalent to that of a substitutional atom about 25 per cent oversize (146). In pure Cu (impurities $<10^{-5}$) Holmes *et al.* (147) find a 17-fold increase in critical shear stress for 10^{19} *nvt* room temperature bombardment. They also report that the magnitude of the yield stress varies with the cube root of the integrated bombardment and with the square root of the absolute temperature, from 4°K. to 150°C., a result which is consistent with a model based on thermally activated Frank-Read sources. Jamison *et al.* have investigated the stress strain curves for bombarded Cu deformed at 78°K.

They find that the flow stress approaches that for unirradiated Cu beyond a strain of about 0.2, since the rate of work hardening is much less for the irradiated material. Presumably, the impedance to dislocation motion is by intersecting slip systems beyond this strain. Similarly Kunz (149) reports that Fe single crystals show much less radiation hardening if they have been previously cold worked. McReynolds *et al.* (150) have studied the effect of neutron bombardment at liquid nitrogen temperature on the critical shear stress of Cu and Al wires. They find a fivefold increase in both cases. They also investigated the annealing of the change in mechanical properties and for Cu found it in striking contrast with the behavior of electrical resistivity. Thus ⅔ of the resistivity annealed below room temperature while there was no recovery of the shear stress, which showed significant annealing only at 350°C., corresponding to the known activation energy for self diffusion. For Al by contrast, the annealing of both resistivity and shear stress were governed by the same activation energy and recovered at about −60°C. Kunz & Holden (151) investigated the flow stress of iron single crystals irradiated at room temperature, and found a recovery process which had an activation energy equal to that for self diffusion in iron (3.1 ev). The authors conclude from this that radiation hardening must be due to agglomeration of defects. Complete recovery can occur only when the precipitated interstitials or vacancies can be swallowed by migration of the vacancies present in equilibrium in the lattice.

A number of studies have been made to investigate the effect of radiation on creep (152). The results of all such experiments appear to be negative, suggesting that creep involves rather long range diffusion processes. This is in line with the theoretical conclusions of Lomer (153) regarding diffusion processes.

The hardening effect of radiation is a matter of some moment in the case of body-centered cubic metals. Radiation produces a rather drastic reduction in impact strength, resulting from a raising of the transition temperature (154). Bruch and coworkers (155) have found that the transition temperature in Mo is raised from −30°C. to +70°C. by an integrated *nvt* of 10^{20}, while similar data are reported on various types of steel by Wilson (156). The transition temperature increase is linear with integrated flux.

Dienes (157) has predicted that elastic constants should increase as a result of damage. In close-packed metals in which the lattice spacing is determined by the very "hard" ion core repulsions, this effect, which arises from the interstitials, should be quite large, amounting in Cu to about a 5–7 per cent increase in shear modulus for each 1 per cent of interstitial content. Unfortunately the observation of this effect is obscured by the occurrence of another increase in Young's modulus which arises from the pinning of dislocations by defects (158). Thompson *et al.* have investigated this pinning effect in some detail using neutron bombardment of very pure copper (159). They find that the effect saturates for very low integrated exposures, of the order of 10^{12} *nvt*, and its magnitude varies with sample, indicating a

high degree of structure sensitivity. The *nvt* values used by these workers, less than 5×10^{17}, were probably so small that the direct effect of interstitials on the shear modulus would have been obscured. Thompson *et al.* (160) have studied both the Young's modulus and decrement as a function of *nvt*, and have analyzed the results in terms of the elastic string model of dislocation pinning proposed by Koehler (158). The theory represents the observations very well, and can be used to estimate the dislocation densities and pinning lengths in the unirradiated material (161).

Changes in shear modulus with irradiation by electrons and also deuterons have been reported by Dieckamp (162). The bombardments used were much greater than in the experiments of Thompson *et al.*, being such as to produce defect concentrations of the order of 10^{-3} to 10^{-4}. They found a decrease in Young's modulus with bombardment at liquid N_2 temperature. This annealed in a rather complicated way (163), changing from a 1.5 per cent decrease at liquid N_2 to a 2.5 per cent increase at room temperature, which subsequently annealed out between 250 and 350°C. The decrease with bombardment is also indicated by electrical resistivity measurements of Bowen *et al.* (164), who show that the temperature part of the resistivity of irradiated Cu can be fitted by a Gruneisen formula with a *lower* Debye temperature than normal Cu. These results are difficult to understand unless it is assumed that interstitials are already aggregated at the temperature of bombardment. Evidently many more experiments at various *nvt* and temperatures will be necessary before all these experimental results can be reconciled and interpreted.

Electrical resistivity.—The changes in electrical resistivity produced by bombardment have already been discussed in the section on the theory of damage. In this section we shall remark only on the comparison of various metals. Such a comparison has recently been made by Redman *et al.*, using neutron bombardment in the very low temperature facility at Oak Ridge (165). The effective temperature was 19°K. They found the following orders of magnitude:

Cu, Au, alpha-brass	1 μohm cm. per 10^{20} *nvt*.
Ni, Al	3 μohm cm. per 10^{20} *nvt*.
Zn, Fe, Co	9 μohm cm. per 10^{20} *nvt*.
Cu_3Au (ordered)	11 μohm cm. per 10^{20} *nvt*.

Work by Wruck & Wert at about 120°K. with charged particles indicates that the change in resistance for Fe is about 3 times larger than for Ni and Co (166), a result which is not in complete agreement with the above. In the case of Cu_3Au the large resistance change is due partly to disordering of the lattice and partly to interstitials and vacancies. This is illustrated by experiments of Brinkman *et al.* (167) on ordered and disordered Cu_3Au bombarded with 9 Mev protons below −100°C. The disordered specimen showed a resistivity increase of 0.65 μohm cm. while the ordered specimen showed 4.25μohm cm. for the same bombardment. The lower figure is at-

tributable to Frenkel defects and is comparable to what may be expected for Cu and Au. The difference of 3.6μohm cm. is then attributed to disordering. Similar effects have been observed in beta brass (168), the change of resistivity for alpha bombardment below -100°C. being about 10 times that for pure Cu. Rosenblatt *et al.* (169) have made an extensive study of resistivity changes in alpha brass, a random solid solution, by neutron bombardment in the low temperature facility at Brookhaven. The resistivity changes observed at 80°K. show an interesting dependence on the composition of the alloy, increasing rapidly with Zn content so that the increase for a 30 per cent alloy is about 3.5 times that for pure copper. This effect is ascribed to the much more effective scattering of interstitial zinc atoms. The resistivity changes recover almost completely when the samples are warmed to room temperature. If the irradiation is carried out at $+50$°C. two interesting new effects occur. By measuring the resistivity as a function of temperature from room temperature down, it is possible to separate the resistivity change into a residual part $\Delta\rho_0$ and a temperature dependent part $\Delta\rho_T$. The residual is *decreased* by irradiation while the thermal part is increased. Both of these effects are attributed to increases in short range order produced by the radiation, apparently due to enhanced diffusion. Equivalent changes are produced by annealing at 190°C. There are complications, however, because $\Delta\rho_0$ saturates with increasing bombardment while $\Delta\rho_T$ continues to increase. Furthermore, $\Delta\rho_T$ anneals differently from $\Delta\rho_0$.

Density.—Tucker & Sampson (170) have pointed out that Frenkel defects in close packed metals should produce a change in lattice constant equivalent to a volume strain of 2.4 p, where p is the fraction of defects. An equivalent density change should also be observed (171). A few attempts have been made to observe the density change produced by bombardment. McDonnell & Kierstead have studied the volume changes produced by deuteron bombardment of Cu at 77°K. (172). The initial rate of expansion was found to be 19 times smaller than predicted. The rate of expansion was 10 times smaller for bombardment at 25°C. There was evidence of annealing even at 77°K. as indicated by a strong trend towards saturation. Although 10 per cent of the volume change annealed below -105°C., 80 per cent of it remained even after annealing to 400°C. These results are strongly suggestive of aggregation of defects. It is interesting to note, however, that the result for 77°K. bombardment is consistent with the number of displacements computed from Overhauser's stored energy measurements (27). The lack of annealing at 400°C. is surprising in view of the fact that self-diffusion alone should produce recovery at this temperature, as evidenced, for example, by the recovery of mechanical properties.

Annealing Characteristics and Identification of Defects.—The recovery of damage in bombarded metals has been studied mostly by means of residual resistivity. The interpretation of annealing experiments on Cu, Ag, and Au, and on Cu_3Au and CuZn has been reviewed by Seitz and Koehler (9, 73), and the situation will be summarized only briefly here. Annealing of deu-

teron damage in Cu, Ag, and Au irradiated at 10° to 15°K. has been reported by the Illinois group (6). Detailed isothermal annealing studies on deuteron bombarded Cu at liquid nitrogen temperature have been reported by Overhauser (173), who has also discussed the annealing of stored energy (27) for the same conditions of bombardment.

The results on pure metals may be summarized as follows. There is evidence for strong annealing at 30° to 40°K. corresponding to an activation energy of about 0.1 ev. This process is much less prominent in Au than in Cu and Ag. In view of Huntington's (174) prediction of an extremely small activation energy for migration of interstitials this low temperature annealing has sometimes been associated with interstitial migration although the nature of the process is not clear, especially in view of the fact that there is essentially no detectable stored energy release associated with this process and those which occur at even lower temperature (175). It may possibly be associated with trapping of interstitials at impurities as suggested by Cottrell & Lomer (176). Some recovery takes place at all temperatures from 30°K. to about −30°C., but is apparently not associated with a well defined activation energy (173). A rather prominent annealing peak occurs at −30°C., associated with an activation energy of about 0.7 ev. This has been variously attributed to interstitial migration (49) and vacancy migration (173). Its isothermal annealing characteristics have been carefully studied by Overhauser and shown to be nearly bimolecular and so probably attributable to mutual annihilation of vacancies and interstitials. Only a very small fraction of the resistance change due to bombardment remains above room temperature. In cold-worked copper, on the other hand, there are two well defined annealing states, one with an activation energy of 0.7 ev, and the other with an activation energy of 1.19 ev (177). Since cold work is believed to produce vacancies preferentially (178) whereas radiation produces vacancies and interstitials in equal numbers, Brinkman *et al.* (167) have suggested that the lower annealing state is due to interstitials and the upper due to migration of vacancies to dislocations and grain boundaries. These authors show that the kinetics of the upper annealing state is that which would characterize migration to sinks, starting very rapidly and trailing off very slowly. Brinkman *et al.* believe this view to be further supported by study of the annealing of bombardment damage in Cu_3Au. The recovery of the resistivity shows two annealing peaks roughly corresponding to those in pure Cu. The high temperature recovery peak near 150°C. is prominent in initially ordered material but not in the disordered. This is interpreted as evidence for the growth of ordered domains in the material partially disordered by radiation, but not in the totally disordered material. Since only vacancy migration can cause ordering, the low temperature annealing peak is ascribed to interstitials. If this general interpretation is correct, it is in disagreement with calculations of Huntington (174) and leaves all the low temperature recovery, which is responsible for 60 per cent of the total resistance change, unaccounted for. This could be attributed to annihilation of

very close pairs, as suggested by Overhauser, but in this case the frequency factor is much too small (179). Furthermore, the absence of stored energy release in the 30°K. recovery process as reported by Blewitt *et al.* fairly well precludes a simple annihilation process.

An alternative view has been proposed by Nowick (180) who suggests that the +150°C. peak is due to vacancies and the −30°C. peak to divacancies, *i.e.*, coupled pairs of vacancies. Bartlett & Dienes (181) have computed that the migration energy for divacancies should be less than that for single vacancies. It is difficult to see how the divacancies could be formed in the first place, however, without an accompanying migration of single vacancies. Furthermore, we must assume that the interstitials somehow migrate and coagulate or become trapped by impurities (176) at low temperatures without annihilating any significant fraction of the vacancies. This might have been barely possible in the experiments of Cooper *et al.*, whose crystals had an impurity content of the order of 3×10^{-4} compared with a maximum vacancy concentration which could not have been less than 10^{-4}. What is needed is a repetition of Overhauser's isothermal annealing experiments with ultra high purity Cu and also Cu of controlled impurity content bombarded at liquid He temperature. Also a more detailed study of stored energy release both below 90°K. and above room temperature would be desirable.

If vacancy migration is responsible for the 150°C. recovery in Cu, then its absence in irradiated Cu is readily understood as due to mutual annihilation of the nearly equal numbers of vacancies and interstitials at lower temperatures. Additional evidence for this view is provided by the work of Li & Nowick (182) who have studied the elastic after effect in an alloy of 17 per cent Al in Cu. This effect, attributable to stress-induced local order (183), is a measure of the product of vacancy concentration times vacancy mobility. It may be characterized by a relaxation time τ which may be expected to vary with vacancy concentration. When the alloy is quenched from a high temperature, τ is initially very short, because of quenched-in vacancies, and increases rapidly as the sample is annealed at temperatures above 200°C. This method is capable of detecting a vacancy concentration of 10^{-5} at 150°C. and yet when the alloy is irradiated with neutrons at liquid N_2 temperature to an *nvt* of 10^{19}, it is found that τ is essentially infinite up to 200°C. and above this temperature takes on the value corresponding to equilibrium vacancy concentration. It must be assumed that substantially all vacancies have been removed by warming to room temperature, and that any remaining damage (about 10 per cent of the total resistivity change in alpha brass, for example) is due to aggregations of defects, such as stacking faults. It seems unlikely that the destruction of vacancies would be less complete in a pure metal such as Cu than in the alloy.

A further confusion is the question of the relative value of the migration and formation energy for vacancies. In alloys, where migration and formation energy can be measured separately by equilibrium and non-equilibrium

studies (184), it is invariably found that the migration energy is 60 per cent of the total activation energy for self diffusion. On the other hand, Kauffman & Koehler (185) have concluded from quenching studies on Au that the migration energy is only 40 per cent of the total, a result which would fit nicely with the 0.7 ev recovery peak in Cu being due to vacancies. However, there is evidence that this peak also occurs in alpha brass. Furthermore, this interpretation would leave the 1.2 ev recovery observed in cold-worked Cu unexplained. These questions apparently cannot be resolved on the basis of present evidence.

Valence Crystals

Crawford & Wittels have reviewed the information available on valence crystals (186). Observations have been mostly on optical properties and X-ray structure with some work on density and on paramagnetic resonance.

Diamond.—Diamond is the prototype valence crystal and as such has received considerable attention. Diamond crystals show lattice expansion and coloration as a result of fast neutron bombardment. The expansion saturates at a fast *nvt* of about 3×10^{20} corresponding to a density decrease of about 4 per cent, and the lattice expansion and density change agree, indicating that the defects formed are Frenkel pairs (186). Levy & Kammerer (187) irradiated diamond powder to 3.8×10^{20} *nvt* in the MTR and found considerably different results. This material turned black, showed a lattice expansion of 2.4 per cent, and the x-ray patterns were those of an amorphous material. Keating (188) has investigated this powdered material in greater detail. From the small angle scattering he has constructed a radial distribution function and in this way studied the numbers of atoms in successive shells. The diamond lattice has the interesting property that the largest interstitial positions have the same size and coordination as normal lattice sites. The interstitial and normal sites together constitute a body centered cubic lattice with half the cube edge of the diamond structure. The damaged diamond lattice thus may be described as approaching the condition of a random array of vacancies and atoms on a body centered cubic arrangement. Keating finds that the observed radial distribution for an *nvt* of 6×10^{20} can be fitted to a picture of this sort. However, as yet is has not been possible to interpret this distribution function in terms of the location of displaced atoms.

Griffiths, Owens & Ward have reported the results of paramagnetic resonance studies on irradiated diamond (189). They find two types of lines, one with an isotropic g-factor of 2 and the other a weaker line with an anisotropic g-factor. O'Brien & Pryce (190) have interpreted the isotropic line as arising from vacancies. They suggest that a vacancy can trap 4 electrons and by Hund's rule will form an $S=2sp^3$ configuration, i.e., ^5S with negligible crystal splitting. They suggest that the interstitials form a non-magnetic configuration $3P_0$, and the weak anisotropic line arises from C_2 complexes, probably having $S=1$ and with their axes oriented in the six (110) direc-

tions. It is noteworthy that the paramagnetic resonance appears only after bombardment with particles capable of displacing atoms, i.e., neutrons or energetic electrons, and the strength of the resonance is proportional to *nvt* and independent of the type of diamond. The concentration of centers is of order 10^{-3} for about 10^{19} *nvt*, which is consistent with the results of Keating (188). The vacancy line could be annealed by heat treatment, but the C_2 line appeared more stable against heating.

Primak, Fuchs & Day (191) have reported stored energy up to 1.2 kcal /mole or 0.05 ev per atom. Annealing phenomena are complex, some properties failing to recover after annealing at 1600°C. Pringsheim (192) has investigated the absorption spectra of diamond irradiated with neutrons. The change consists mainly of a general enhancement throughout a broad spectral region. Similar results have been reported by Dugdale (193). The most extensive optical investigation is that of Ditchburn *et al.* (194) who have investigated both electron and neutron bombarded diamond. They find an enhanced absorption extending in a broad band from 1.65 to 2.50 ev and considerably stronger enhancement in the ultraviolet between 3 and 4 ev. At high resolution and at liquid N_2 temperature the absorption shows considerable structure. Very little progress has been made in interpretation of the results. Presumably, in analogy with Ge and Si, the ultraviolet band is associated with excitation of electrons from the vacancy into the conduction band while the weaker absorption at about 2 ev is associated with an excited state of the vacancy and possibly also with excitation from an interstitial into the conduction band. This would suggest that the main defect levels are symmetrical in the band gap as in Si.

Quartz.—Quartz has also been extensively studied, both as crystal quartz and as fused silica. Crystal quartz is changed to the amorphous form by heavy particle bombardment, this decrease being accompanied by a density decrease of 15 per cent which saturates at about 10^{20} *nvt* (195). At a relatively early stage ($nvt \times 10^{19}$) the c/a ratio is lowered below that which the high temperature modification β-quartz exhibits at 1000°C. Consistent with this quartz bombarded to 5×10^{19} *nvt* shows no evidence of the latent heat characteristic of the α-β inversion when the crystal is heated through the transition. On the other hand, the bombarded material can be transformed back to polycrystalline α-quartz by prolonged annealing at 930°C., just below the inversion temperature.

The magnetic properties of crystalline quartz are also of considerable interest. Stevens (196), by static susceptibility measurements, has found evidence for a paramagnetic center, which builds up to a maximum concentration of about 10^{-3} at an *nvt* of 5×10^{19} and then falls off. This center has been further studied by paramagnetic resonance by Weeks (186), who found an exceedingly complex spectrum with a multiple line structure. At higher exposures the complexity disappears and the line becomes isotropic at an *nvt* of 3×10^{20} in agreement with the complete amorphism indicated by x-ray results. Griffiths *et al.* (189) found similar results at the lower exposures.

However, O'Brien & Pryce (190) have interpreted these results in terms of an Al substitutional impurity in the lattice, the complexity of the line arising from electronic hyperfine iteration with the $I=5/2$ Al nucleus. It is noteworthy that Griffiths *et al.* also observed the resonance line with x-radiation, which they interpreted in terms of the reaction $Al \rightarrow Al + e$, the neutral Al being paramagnetic. The evidence indicates fairly clearly that the resonances observed by the Oak Ridge group are to be ascribed to bombardment produced defects in the pure crystal and are not caused by impurities. Two distinct lines with g-factors greater than and less than the free electron value are ascribed to holes and electrons, respectively. More work is needed to relate the work of the Oxford and Oak Ridge groups.

Fused Silica.—Fused silica has also been investigated. Perhaps the most interesting results are those obtained on Corning "pure" silica glass, which is highly resistant to coloration by ionizing radiation. This permits observation of the ultraviolet bands at 2150Å and 2500Å which are developed by the radiation (186, 197). The efficiency of various types of radiation varies widely. Soft x-rays produce a band which saturates at an absorption coefficient more than ten times smaller than that produced by fast neutrons. The absorption in the 2150Å band builds up and decays in very close correspondence to the paramagnetic resonance absorption and probably originates from the same center. Not much progress has been made towards developing a detailed model.

In less pure fused quartz, a complex optical absorption in the visible is developed (198). Some progress has been made in identifying the bands with impurities known to be present.

The contrasting effects of gamma and neutron irradiation are shown even more in the case of α-Al_2O_3 (199). Gamma radiation produces two weak bands at 2300 Å and 4000Å which saturate at about 3×10^4 roentgens. Neutron irradiation by contrast produces a strong band at 2040Å and a weaker band (one-fifth as strong) at 2600Å. The 2040Å and 2600Å grow accurately together as a function of *nvt*, and include a linear and a saturating part. Exposures were in the range 10^{16}–10^{17} *nvt*. The absorption bands are undoubtedly associated with displaced atoms, and one must conclude the gamma ray bands are associated with impurities.

Studies have been made on many other natural crystals of the valence type. In general, one may say that the susceptibility to heavy particle bombardment damage, as indicated by x-ray analysis, varies directly with the degree of covalent binding. Space does not permit discussion of all these data, which mostly are not yet at a stage where any interpretation is possible (186).

Alkali Halides

The defects produced in alkali halides by ionizing radiation have been investigated for many years, especially as regards their optical spectra (76). In general the centers produced by heavy particles are similar. In the pres-

ent brief summary the discussion will be limited to those effects which are largely unique to heavy particle damage. Such effects have been reviewed recently by Smoluchowski (200). We will also confine our attention to the alkali halides, since these are the best understood.

Lithium Fluoride.—LiF is a convenient material for study because slow neutron irradiation results in "fissioning" of the Li^6 nucleus into helium and tritium with a kinetic energy of nearly 5 Mev divided between them. As a result the damage produced by a given flux is an order of magnitude greater than would be produced by the same fast flux, and the material is hence suitable for study in low power research reactors.

Extensive x-ray structure investigations have been made on LiF (201, 202, 203) exposed in thermal reactors. The measurements include small angle scattering (203), diffuse scattering (202, 203), and studies of lattice parameter and density (201, 203). Thermal fluxes used range from 10^{17} to 10^{19} *nvt.*

Up to about 10^{17} *nvt* the lattice constant increases linearly with total dose, about 0.22 per cent for 10^{17} *nvt* (201) with little or no evidence of line broadening. The density change agrees within 6 per cent with the lattice expansion, indicating that the defects must be largely of the Frenkel type.

For higher doses the lattice parameter passes through a maximum and both line broadening and small angle scattering begin to appear. For a dose of the order of 10^{19} *nvt*, the lattice parameter returns practically to its original value. Very strong diffuse scattering is found, with prominent streaking in the (100) direction in reciprocal space between the origin and the (200) reciprocal lattice point.

The scattering results have been interpreted in terms of the coalescence of both interstitials and vacancies on definite lattice planes. The interstitials are believed to form platelets about 50 Å in diameter, and these produce local distortions of the structure in the (100) direction corresponding to a 30 to 50 per cent expansion of the lattice planes locally around the interstitial precipitate. The vacancies are thought to coalesce into flattened cavities with a radius of gyration of about 32 Å. There is also evidence for some disoriented regions which anneal out rather more readily than the defect precipitates.

A remarkable observation is that even after the most severe damage the complex defect structure seems to be eliminated by annealing with no tendency towards recrystallization (203).

Jen *et al.* have observed paramagnetic resonance in LiF bombarded either with x-rays or with protons (204). The heavy particle bombardment gives a greater line breadth by a factor of 2, presumably due to dipolar interactions between defects resulting from clustering along the primary track. Further evidence for interaction is that the various optical absorption bands are displaced slightly from their "normal" positions, even for bombardments as low as 10^{14} protons per $cm.^2$ (205) in NaCl and KCl.

Electrical and Optical Effects in NaCl and KCl.—Mapother (206) showed that self-diffusion in NaCl was reduced by exposure to x-rays. One might expect similar effects for the ionic conduction, and such were observed by Smoluchowski and his colleagues (207). They bombarded KCl with 400 Mev protons and observed the ratio of its resistance to that of a normal crystal as the two were warmed together. A 60-fold increase in resistivity was observed in NaCl. The ratio of irradiated to unirradiated resistivity reached this peak value at 120°C. and gradually fell to unity as the crystal was further warmed to 400°C. Very similar results were observed in KCl irradiated with gamma rays, the maximum increase being over 100-fold. A second peak in the resistivity ratio at 250°C. is observed in heavily bombarded NaCl.

Two explanations of this rather remarkable effect have been put forward. One suggestion, which is in accord with Varley's (79) mechanism, involves trapping of positive ion vacancies, ordinarily the conducting defect, by excess negative ion vacancies generated during the irradiation. The effect only becomes operative when the temperature is high enough for the positive ion vacancies to migrate. The isothermal annealing of the resistivity increase (207) indicates that it disappears monomolecularly with an activation energy characteristic of negative ion vacancies. Mapother originally suggested that the resistivity effect was due to trapping of holes at positive ion vacancies, but this seems to be ruled out by the well-known thermal instability of V-centers. Optical experiments also indicate that most of the centers formed are F-centers, which would further tend to rule out hole trapping (207).

According to stored energy measurements (208) recombination of vacancies and interstitials in NaCl is believed to take place at about 200°C., before the second peak in resistivity ratio. This second peak must therefore be ascribed to trapping of positive ion vacancies by larger clusters.

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APPLICATIONS OF OXYGEN ISOTOPES IN CHEMICAL STUDIES[1]

By Henry Taube

Department of Chemistry, University of Chicago, Chicago, Illinois

The choice of the general subject of this article requires no apology, but the choice of the specific material to fit to the space allotted does. The limitations and research interests of the reviewer were a governing factor in making the selection, but also the consideration entered that the effects of the major omissions are tempered by other recent reviews in which much of the material omitted is discussed. Thus the important work in geochemistry involving oxygen isotopes is discussed in excellent reviews by Dole (1), and more recently by Craig & Boata (2). The work on heterogenous systems is discussed more completely in the article by Dole. It is hoped in this connection that a general article on the application of isotopes in the study of heterogeneous catalysis, including work with oxygen isotopes, will be forthcoming. In the subjects chosen for review, an effort has been made to be complete; and earlier literature as well as the more recent has been referred to. When work which is within the scope of the subject matter is not cited, it is because of oversight or because the work is dealt with in a more general article by the same author.

The present article stresses the application of oxygen isotopes to the study of chemical phenomena in homogeneous solution. Much of the work exploiting oxygen isotope effects has been done for such systems. For reactions of oxygen-containing substances in an oxide-labile solvent such as water, isotope studies can answer important questions which no other method can answer as directly; and they have been of great help in defining the nature of the solvent and of the substances dissolved in it, and in exposing the mechanisms of reactions in solution.

Techniques

In the work reviewed, mainly the oxygen isotopes of mass 18 and 16 come into question. The mass spectrometric method of measuring isotope ratios has almost completely supplanted that depending on the determination of density differences in water. With care, commercial instruments can measure differences to a precision of 1 part in 1000 in O_2 or CO_2 of normal oxygen-isotopic composition. With specially designed instruments (3), a precision of one part in approximately 10,000 on CO_2 can be attained.

When the oxygen is available as water, an accurate method (4) of isotopic assay is equilibration of CO_2 with the H_2O, followed by measurement of the isotopic ratio in the CO_2. Dostrovsky & Klein (5) have described the high temperature equilibration of CO_2 and H_2O, and have derived an equation

[1] The survey of literature pertaining to this review was completed in January, 1956.

relating the original isotopic composition of the water to the isotope ratio measured in CO_2. When only very small samples of H_2O are available, the complete oxidation of the sample to O_2 can have advantages. However, after expending considerable effort to develop such a method using BrF_3, Baldwin & Taube (6) abandoned it and turned to equilibration with CO_2, determining the amount of water by an isotopic dilution method.

A major problem in oxygen tracer work, already referred to for the special case of water, is that of converting the oxygen to a form amenable to mass-spectrometric analysis. The problem is both particularly important and particularly difficult as it is met with among organic compounds. The reduction of such compounds by H_2 has been used (7, 8, 9), as well as oxidation by a known excess of O_2 (9), but these methods have not been developed for precision use. Doering & Dorfman (9) preferred to adapt the Schutze-Untersaucher method of oxygen analysis to the problem of isotopic analysis. The compound to be analyzed is heated at 1120°C. over a carbon catalyst to yield CO, and this is oxidized to CO_2 with I_2O_5. Bender and Dewey (10) have reported a considerable blank using this method, i.e., CO_2 in excess of that expected from the oxygen in the original sample. Jensen and coworkers (11) have been able greatly to reduce the blank by substituting a platinized carbon catalyst (12) thus permitting them to lower the temperature of pyrolysis to 930 ± 5°C. Anbar *et al.* (13) have shown that it is possible to equilibrate oxygen of a variety of organic substances directly with CO_2 at 150–200° by using a trace of H_2SO_4 as catalyst.

Sulfates (14) have been handled by reducing $BaSO_4$ with C using induction heating to avoid the exchange with glass parts of the system, phosphates by reduction with C (15) or by removing H_2O from KH_2PO_4 (16) by heating the salt. Anbar & Taube (17) have shown that oxygen is released from Ag_2SO_4 and Ag_3PO_4 when they are strongly heated (Pt crucible in an induction furnace). While release of oxygen is not complete and fractionation effects will interfere slightly, this simple method may be preferable in many applications to a method involving reduction with carbon, which also introduces problems. A large number of inorganic oxygen compounds respond to fluorine or halogen fluorides. ClF_3 and F_2 have been used in getting oxygen from silicates (18), and BrF_3 has been found useful in getting O_2 from a variety of oxides (19). Sheft *et al.* (20) have developed procedures for the direct analysis of oxygen in some organic and inorganic compounds using $KBrF_4$ and BrF_2SbF_6 as fluorinating agents. These methods may lend themselves to use also in the determination of oxygen isotope ratios in some compounds.

With increasing interest in infrared and nuclear magnetic resonance techniques, and with commercial instruments of high quality becoming available, it seems likely that these techniques will soon be applied to the routine determination of oxygen-isotope compositions. The natural abundance of O^{17} is only one-fifth that of O^{18}, so to give the NMR technique full power, higher levels of enrichment will be required. The chemical shifts for O^{17} are large (21, 22)—thus oxygen bound in H_2O is readily distinguishable from oxygen in acetic acid or hydrogen peroxide. This has the important consequence

that in using the NMR technique for isotopic analysis, in many cases it will be unnecessary to convert the oxygen-containing substance to a new form. It has the still more important consequence that the chemical shift can be exploited in solving numerous important problems of structure, equilibria and rates for oxygen containing molecules, of the kind referred to later, and will greatly extend the scope of oxygen isotopic applications. Such applications have only been begun, but rapid progress can be expected when oxygen enriched in O^{17} becomes available.

Little work has been reported using the infrared or Raman techniques for the analysis of oxygen isotopes. The important use of these as of other isotopes in the analysis of rotational and vibrational spectra is not reviewed.

Nature of the Solvent

Wang *et al.* (23, 24, 25, 27) and Graupner & Winter (26) have measured the diffusion rate of H_2O^{18} in water and compared it with the rates for HDO and HTO. The respective diffusion coefficients, D in 10^{-5}cm.^{2} sec. at 25°C., are (25): 2.66 ± 0.12, 2.34 ± 0.08 and 2.44 ± 0.06. A direct conclusion based on the comparison is that the chain transport of H is not an important process for the diffusion of this element. This conclusion is consistent with Eigen's (28) measurement of the rate of dissociation of H_2O as 2×10^{-5} sec.$^{-1}$. The activation energy for diffusion: 4.6 kcal mole^{-1}, is the same as those for dipole orientation and for viscous flow, implying the same kind of molecular process as being rate determining for all (25). The constancy of the ratio $D\eta/T$ over the temperature range 0° to 55°C. suggests that a single species is diffusing, and supports the model for the structure of water as a quasi-crystalline aggregate, rather than as a mixture of polymeric species.

This conclusion is supported also by calculations Feder (29) has made of the isotopic fractionation of oxygen (or hydrogen) in distillation of water. Some vibrational frequencies in the liquid can be recognized as internal modes of the molecule, with frequency slightly shifted from the values for the gas. If these assignments are used, if the frequency at ca. 175 cm.$^{-1}$ is ascribed to hindered translation, and if the poorly resolved absorption in the range 320–1000 cm.$^{-1}$ is ascribed to hindered rotation with characteristic frequencies of 475, 590 and 755 cm.$^{-1}$, the isotope fractionation of water on distillation can be accounted for. If the changes in frequency with temperature of some of the modes as indicated by infrared and Raman data are allowed for, and for others are estimated with these as guides, the fractionation data (30, 31) as function of temperature are also accounted for over the whole range. The external modes in response to interactions in the liquid favor H_2O^{18} being bound more strongly than H_2O^{16} in the liquid, but the change in frequency for the internal modes on condensing the vapor offsets their effect to some extent.

In this connection it is interesting to note that substitution (32) in the CH_3 part of CH_3OH results in only minor discrimination effects, while in the OH part, which presumably is the seat of the strong intermolecular attractions, it produces a large response.

Teis & Florenskii (33) have observed concentration of O^{18} in ice when it is formed from water.

Oxygen Hold-Back by Dissolved Substances

It is important for tracer applications to know whether reactants and products exchange oxygen rapidly with solvent. In addition to such specific application, the information on hold-back is important in defining the nature of the dissolved substance.

Inorganic molecules and anions.—The following inorganic substances have been found to exchange oxygen slowly enough in neutral or alkaline aqueous solution so that hold-back under these conditions is easily observed (oxyanions become more labile to exchange in acid solution—see section on Rates and Mechanisms of Exchange dealing with inorganic anions): ClO_4^- (34, 35), ClO_3^- (34, 35, 36), ClO_2 (14), ClO_2^- (14), ClO^- (37), BrO_3^- (35), BrO^- (37), O_2 (38), O_3 (39), H_2O_2 (40, 41), $SO_4^=$ (34, 35, 38, 42, 43, 44), $SO_3^=$ (35, 45), $S_2O_3^=$ (45), $SeO_4^=$ (35), NO_3^- (34, 35, 36), NO_2^- (35), $N_2O_3^=$ (47), N_2O (48), $N_2O_2^=$ (47), phosphates (34, 42, 44, 46, 49), $H_2PO_3^-$, $H_2PO_2^-$ (50), $CO_3^=$ (35, 46, 51), Et_3SiOH (52), Φ_3SiOH (52), $CrO_4^=$ (34, 35, 45), MnO_4^- (35, 45). For these substances it is demonstrated that oxygen atoms as shown in the formulas are held distinct from solvent oxygen, and thus for any of them forms in which contained oxygen becomes equivalent to solvent oxygen are ruled out as being readily accessible (e.g., for Cl(I): $Cl(OH)_2^-$ or $Cl^+ + 2OH^-$). The species IO_3^- (14, 35), $SeO_3^=$ (35), $AsO_4^=$ (35, 46), AsO_2^- (35), borates (34, 35), $MoO_4^=$ (35), $WO_4^=$ (35), and silicates (34) have been found to exchange within the time of isotope sampling, but since stringent conditions were usually used (100°C. and long contact time or both), these ions deserve further study.

Organic molecules and anions.—Hold-back of oxygen by a variety of organic groups has been demonstrated. They include RCO_2^- (4, 53, 54, 55, 56, 57), amide (55, 59, 60), ester (4), RR'C=O (4), aldehyde (4, 54, 55, 56), ROH (4, 54, 56, 57, 61), ΦNO_2 (57), ϕOH (56), sugars (56, 62), $(NH_2)_2C{=}O$ (4), cellulose nitrate (58). Adjacent groups can labilize oxygen in the functional groups, but in ordinary settings the organic groups referred to retain oxygen distinct from that of the solvent for hours at room temperature.

Inorganic oxy-cations.—The first cation to be studied by the oxygen-exchange technique was UO_2^{++}. Crandall (63) showed that U(VI) holds back two oxygen atoms, a result consistent with the formula above, but not with $U(OH)_4^{++}$, which would otherwise also appear to be a reasonable formulation.

Aquo cations.—Hold-back of oxygen by cations is not limited to that bound as oxide, but can be observed also for that bound as H_2O. Hunt & Taube (64, 65) found that Cr(III) holds back six water molecules, thus justifying the formula $Cr(H_2O)_6^{+++}$ as a more detailed description of Cr_{aq}^{+++}. The mixed aquo complexes $Co(NH_3)_5H_2O^{+++}$ (66) and $Co(NH_3)_4(H_2O)_2^{+++}$ (67) have similarly been characterized as species having identity in solution. Exchanges of H_{aq}^+, Al_{aq}^{+++}, Fe_{aq}^{+++}, Th_{aq}^{++++} (65), Co_{aq}^{+++}, Co_{aq}^{++} (68), Cr_{aq}^{++}, Zn_{aq}^{++} (69) are found to be complete within one minute at 25°C. An

adaptation of a flow technique to water exchange reactions promises to bring certain of the labile aquo ions also within the scope of the exchange method (6). Using the new technique hold-back of water by Al_{aq}^{+++} has been established. The half-time for exchange is greater than 2×10^{-2} sec and the formula $Al(H_2O)_6^{+++}$ is indicated.

Bridge-oxygen compounds.—There is a good possibility that the exchange method can be used to distinguish bridge oxygens in polynuclear ions from water which is singly bound (65), but this potentiality has not been exploited for aquo ions. It has however been used in an analogous problem in protein chemistry. Mears & Sobotka (60) have used the exchange technique to distinguish oxygen in free carboxyl groups from other oxygen bound in the protein. The oxygen in the carboxyl group exchanges more rapidly than that in amide positions.

Equilibrium hold-back.—In the foregoing paragraphs, only the kinetic hold-back of oxygen by dissolved ions was discussed. The equilibrium hold-back of one isotope in preference to another also bears on the problem of the nature of the interaction of ions and water, and provides a tool for its study. For cations which exchange bound water with the solvent slowly, a sensitive direct measure of the equilibrium fractionation by analyses in the aquo cation is possible. For the reaction.

$$Co(NH_3)_5H_2O^{16+++} + H_2O^{18} \rightleftharpoons Co(NH_3)_5H_2O^{18+++} + H_2O^{16},$$

the equilibrium constant at 25°C. is found to be 1.019_5 (66).

For exchange-labile cations, direct measurements of this kind are not possible, but the effects which salts exert on the fugacity ratio H_2O^{16}/H_2O^{18} can be measured. The effects include those caused by exchange in the first sphere of coordination of the cation as well as those which may be caused in the outer sphere water or by the anion. Salts containing cations of high ionic potential, e.g. Al^{+++}, Mg^{++}, H^+, increase the fugacity ratio (29, 70, 71), and the effect is linear in the molality of the salt over a considerable range of concentration. The effects are to a first approximation independent of the nature of the anion, and are very small for a salt such as $Co(NH_2CH_2CH_2NH_2)_3Cl_3$, the cation of which has the first sphere of coordination fully occupied. It seems reasonable to attribute the changes in the fugacity ratio H_2O^{16}/H_2O^{18} to the equilibrium discrimination exerted in the first sphere of coordination of the cation, particularly because the magnitude of the effect for Al^{+++} is in reasonable accord with that expected on this basis in comparison with the equilibrium constant measured for water exchange in $Co(NH_3)_6H_2O^{+++}$. A puzzling phenomenon, however, is that with ions of low ionic potential, e.g., K^+, Cs^+, and NH_4^+, the fugacity ratio H_2O^{16}/H_2O^{18} is decreased (29, 71).

Rates and Mechanism of Oxygen Exchange Between Dissolved Substances and Water

The reactions considered in this section may be regarded formally as classes of substitution reactions, to which a section of this review is devoted; but because the results obtained in their study have an important bearing

also on oxidation-reduction reactions, they are discussed separately in this section. The oxygen-exchange behavior of inorganic anions, of organic functional groups, and of aquo cations is considered.

Inorganic anions.—Mills & Urey (51, 72) have made a thorough study of the exchange of $CO_3^=$ with water. For the pH range in which CO_2 or HCO_3^- are the dominant forms of C(IV), the activated complex has the composition H_2CO_3, and the rate of exchange is identical with that measured by direct chemical means for the rate of hydration of CO_2 or for the decomposition of H_2CO_3. In alkaline solution, the path involving the activated complex of composition HCO_3^- becomes dominant, and the rate is the same as that measured independently for the reaction $HCO_3^- = CO_2 + OH^-$. Catalysis of the exchange reaction between CO_2 and H_2O has been studied by Reid & Urey (73).

The rate of exchange of nitrate ion and water increases sharply with acidity (74), and is catalysed by nitrous acid (75). The circumstance that the rate of exchange is equal (76) to the maximum rate of nitration of reactive organic substrates shows that a common intermediate is involved, and this may reasonably be taken to be NO_2^+.

Careful and thorough work by Hoering (77) on the exchange of $SO_4^=$ and water shows the composition of the activated complex to be H_2SO_4, and suggests that solvent H_2O is not contained in it. Thus the products formed in the rate determining step are probably SO_3 and H_2O.

Anbar & Taube (78) found the composition of the activated complex for the exchange of NO_2^- and water to be $2H^+ \cdot NO_2^-$; but Stedman & Bunton (79) report the composition $2H^+ \cdot 2NO_2^-$. In view of the opinion expressed by Stedman and Bunton that one of the NO_2^- ions in the activated complex may be replaced by other anions, Anbar & Taube (80) have repeated some of their experiments with all anions other than NO_2^- kept at low concentration, and still find the activated complex to have the composition $2H^+ \cdot NO_2^-$. The rate of the NO_2^-—H_2O exchange is equal to the maximum rate of the reaction of NO_2^- with H_2O_2, and both follow the same rate law if H_2O_2 concentration is high (78). The comparison proves that both reactions wait for a common intermediate which may be NO^+.

In the anion systems described thus far, the evidence suggests that exchange takes place by S_N1 mechanisms (81). In each case the intermediate of lower coordination number which is formed can be stabilized by electron rearrangement. The oxygen exchanges for the anions BrO^- and ClO^- (37), however, appear to proceed by S_N2 paths (81). The dominant terms in the rate laws for alkaline solution are k(HOBr) and k(HOCl). A kinetic term of similar form appears in the reaction of HOCl with phenol (82), but the coefficient is much smaller than for the exchange of ClO^- and water. If the rate of the reaction of HOCl with phenol is taken to measure the rate of formation of $Cl^+ + OH^-$, it follows that the exchange reaction must proceed by direct substitution in HOCl. The exchange of ClO^- with water is catalyzed by Cl^-, and of BrO^- with water by Cl^- and Br^- (37). The rate law term for

the catalyzed exchange of ClO^- is $k(HOCl)(Cl^-)$. The direct and simple explanation that this term corresponds to the mechanism

$$Cl^- + HOCl = Cl_2 + OH^-$$

is set aside by the observation (83) that the exchange between ClO^- and Cl^- can take place at a smaller rate than the Cl^- catalyzed oxygen exchange of ClO^- with water.

The exchange of ΦCOOH with water (84) proceeds by a path described by the rate term k(ΦCOOH) (H^+). Definite conclusions about mechanism are not possible in this system, although an S_N2 process is indicated because of the kinetic similarity of the exchange reaction to the hydrolysis of related esters, the latter being presumed to occur by H_2O attacking the carboxyl group. The kinetic similarity extends to the effects of substituents on the rates of the two reactions (85).

For all of the anions mentioned the activated complexes for oxygen exchange with water have been shown to contain H^+. The function of H^+ presumably is to strip off $O^=$ in an S_N1 mechanism, or to withdraw electrons from the center undergoing nucleophilic attack in an S_N2 mechanism. The labilization by H^+ of anions for exchange, at least in a qualitative way, has been noted in numerous other cases: among them ClO_3^- (14), $PO_4^{\equiv}$ (44), $CrO_4^=$ (86). The claim made in the earlier literature (42, 49) that substitution by OH^- in the anion provides a path for exchange in no case has been substantiated by later, more thorough work.

Organic functional groups.—The exchange between carboxylate ion and water has been referred to in the preceding section. The exchange of oxygen between acetone and water is subject to catalysis by OH^-, and to general acid catalysis (4). The oxygen exchange reaction is much more rapid than is the enolization reaction under the same conditions. Catalysis by H^+ and OH^- is reported also for the exchange of benzaldehyde (54) and acetaldehyde (59) with water. The exchange of benzil and water is catalyzed by OH^- and takes place more rapidly than does the rearrangement to benzilic acid (87).

Bender and coworkers (10, 88) have shown that in the hydrolysis in alkaline solution of ethyl, isopropyl, *t*-butyl benzoate, and of methyl *2,4,6*-trimethylbenzoate, exchange of the carbonyl oxygen of the ester with water takes place. These observations are consistent with the view that in the hydrolysis of esters, an intermediate containing an additional oxygen atom is formed. Adopting this view, the ratio of the rates at which the intermediate decomposes to hydrolysis and exchange products can be calculated. For ethyl benzoate this ratio is 4.8 for the alkaline path and 5.2 for the acid catalysed path; for benzamide (89) the ratios are 0.28 and >374. No exchange is observed in the hydrolysis of methyl β-phenyl propionate catalyzed by α-chymotrypsin (90).

The exchange of *sec*-butyl alcohol with water in dilute $HClO_4$ takes place at one-half the rate of racemization, thus proving that inversion takes place in every substitution act. This is ordinarily taken as evidence of an

S_N2 mechanism, with inversion of configuration for each act of direct substitution. Nevertheless, taking account of the variation of rate with acidity, which follows the Hammett acidity function rather than (H_3O^+), Bunton, Konasciewicz & Llewellyn (91) reach the conclusion that H_2O does not enter the activated complex, and make the interesting suggestion that the entering water molecule is blocked by that leaving from making a successful approach on the same side. Dostrovsky & Klein (92) have observed that for *t*-butanol in acid, the oxygen-exchange reaction much exceeds the elimination reaction in rate. The *t*-butyl cation is suggested as a common intermediate for both reactions. Exchange and decomposition rates were compared (93) also for *n*-butyl and neo-pentyl alcohols, and conclusions about mechanisms were drawn.

Aquo cations.—The study of the rate of exchange of cation-bound water and solvent water offers some hope of contributing to an understanding of the mechanism of substitution in complex ions. The scope of subject is wider than might at first sight seem to be the case, for the majority of complex ion substitution reactions in water proceed by intermediate aquation, and in all such cases the problem is reduced to the difficult one of describing the role of the solvent as reactant or product.

In principle, S_N1 and S_N2 processes could be distinguished on this basis: in an S_N1 process, water exchange and complex ion formation compete for a common intermediate; in S_N2 processes, the reactions are completely independent. In practice, no dependable conclusions have yet been reached applying these principles. While in no case has the rate of complex formation for an aquo ion been found to exceed the rate of water exchange, this may only mean that water is uniquely reactive, perhaps by virtue of molecular properties as well as abundance. The attempt by Rutenberg & Taube (66) to discover the mechanism of water exchange for $Co(NH_3)_5H_2O^{+++}$ by studying the competition between water exchange and sulfate complex formation failed because $Co(NH_3)_5H_2O^{+++}$ and $SO_4^=$ associate to an outer-sphere complex ion.

In the following, the work on the kinetics of aquo cation-water exchange which has been done is summarized without further reference to the general question of the mechanism of the substitution reaction. $Cr(H_2O)_6^{+++}$ has been studied most intensively. The values recorded by Hunt & Taube (65) for the specific rate of the exchange reaction for this cation were checked in later work by Plane & Taube (69) which extended the range of conditions studied. Hunt & Plane (94) developed a method for direct analysis of the aquo cation (rather than depending only on the isotope analysis of solvent), and have made the most refined measurements of rate and tests of rate law. Their work yielded for the specific rate of exchange at 27.5°C., 2.0×10^{-4} min^{-1}, and 27.6 kcal/mole for the energy of activation. Furthermore, they showed the claim by Plane & Taube (69): that the rate is first order in anion concentration, to be without substance. In fact, it is remarkable that the rate in the presence of ClO_4^- changes by less than a factor of 2 over the range of ionic strength from 0.4 to 7. Mackenzie & Milner (95) have made

measurements for concentrated solutions of $Cr(H_2O)_6Cl_3$. The rates and activation energies are substantially the same as those for perchlorate media.

The specific rate for the exchange of $Co(NH_3)_5H_2O^{+++}$ and water (66) is 4.8×10^{-4} min^{-1} at 25°C., and is unaffected by moderate changes in acidity or ionic strength.

The exchange of $Cr(H_2O)_6^{+++}$ and H_2O is catalyzed (69) by Cr^{++}. The catalysis evidently takes place by an electron exchange between Cr^{+++} and Cr^{++}, and the specific rate for the electron exchange calculated from the catalytic effects agrees with that measured directly using radioactive chromium (96). $Cr(H_2O)_6^{+++}-H_2O$ exchange is induced (69) by Ce(IV). In this system, catalysis by unstable oxidation states of Cr, intermediate between +3 and +6, may be involved.

Miscellaneous Exchange Reactions

An interesting comparison relating to oxygen exchange between solute and solvent is provided by the observation of Nakata (97): that exchange of oxygen between SO_2 and SO_3 is rapid; and Huston's (98) observation: that sulfur exchange does not take place between these species under the same conditions. Huston has pointed out that the observations are not incompatible, for sulfur exchange requires transfer of O, essentially an oxidation-reduction reaction, while oxygen exchange can take place by an acid-base reaction, e.g., $SO_2+SO_3=SO^{++}\cdot SO_4^{=}$.

H_2O_2 exchanges with O_2 in alkaline solution, but not in acid (99).

The scrambling reaction:

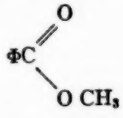
$$O^{18}O^{18} + O^{16}O^{16} \rightleftharpoons 2\ O^{18}O^{16}$$

has been studied by Johnston & O'Shea (100) and by Ogg & Sutphen (101, 102, 103), with results not wholly in agreement. There is agreement, however, that the uncatalyzed reaction is slow. Ogg & Sutphen (103) observed catalysis by O_2, and observed furthermore that the scrambling is more rapid than the O_2-O_3 exchange. They suggest atomic oxygen as the catalytic agent:

$$O^* + O_2 \rightleftharpoons OO^* + O.$$

The exchange is catalyzed also by Cl and by N_2O_5. The scrambling reaction can be a useful tool in studying the action of heterogeneous catalysts, and has been so applied by Winter (104).

Wiberg (105) has shown that oxygen interchange within the molecule



does not become rapid until a temperature of ~400° is reached.

Applications in Studying Mechanisms of Substitution Reactions

A very important application of oxygen isotopes is in determining the point of rupture in a hydrolytic reaction such as:

$$ROR' + H_2O \rightarrow ROH + R'OH$$

or its reverse. In some cases, other evidence on this important feature of mechanism may have been adduced, but in many, a tracer experiment is the only evidence forming the basis for a conclusion. In any case, it provides the most direct evidence which can be obtained on the point in question; and in many systems, ranging from O shared between carbons to O shared between metal and non-metal atom, it has yielded unambiguous conclusions.

Reactions involving organic esters.—The classical experiment by Polanyi & Szabo (53) is an appropriate introduction to the field. These pioneer workers showed that in the alkaline hydrolysis of amyl acetate, substitution takes place at the carbonyl rather than alkyl carbon. Roberts & Urey (106) showed that in the esterification of benzoic acid by CH_3OH in acid, again the alkyl-oxygen bond is preserved. Datta, Day & Ingold (42) showed that the methyl-oxygen bond remains intact in the alkaline hydrolysis of methyl succinate.

Olson & Hyde (107) found an interesting change in the mechanism of hydrolysis of β-butyrolactone with acidity. In alkaline and acid solution, substitution occurs in the carbonyl group, but in neutral solution where the H_2O path is dominant, alkyl-oxygen fission is observed. Acyl-O cleavage is reported by Long & Friedman (108) for the acidic and basic hydrolysis of α-butyrolactone also. Bunton & Wood (109) found a change in mechanism of hydrolysis of *t*-butyl acetate with solvent composition: in water the reaction takes place almost entirely by alkyl-oxygen fission, but in 70 per cent dioxane only 36 per cent of the reaction takes this course. Alkyl-oxygen fission (110) is the principal mode of hydrolysis of triphenyl methyl acetate under all conditions of acidity. It is interesting to note that in this reaction there is very little entry of solvent oxygen into the acid anion, as would be the case if the carbonyl oxygen underwent exchange during the hydrolysis (88).

Some work has gone into the study of the mechanism of the enzymatic hydrolysis of esters. Sprinson & Rittenberg (111) reported acyl-oxygen fission in the action of chymotrypsin on carbobenzyloxy-L-phenylalanine, Bentley & Rittenberg (112) in the esterase hydrolysis of acetylcholine or ethyl acetate. Stein & Koshland (113) had already reported the same conclusion for the enzymatic hydrolysis of acetylcholine, had noted that the same type of rupture occurs for the reaction in alkaline solution, and have pointed out in addition that there is no exchange of acetylcholine with water during hydrolysis.

In the formation of xanthate from C_2H_5OH or cellulose and CS_2 in alkaline solution, the oxygen of the organic group appears in the product (114).

A reaction somewhat related to those considered, although it does not involve ambiguity with respect to the role of solvent, is that of ethyl alcohol with acetic anhydride. Dedusenko & Brodskii (115) have shown this to occur with the alcohol oxygen appearing only in the ester, none in the acid which is also formed.

Reactions involving inorganic functional groups.—A somewhat newer and

less completely charted area in the field of reactions of the type under discussion is that in which non-carbon atoms share the oxygen involved in the hydrolysis. Anbar *et al.* (116) have made a survey of some substances in this field: a group of esters of inorganic acids. They report that *t*-butyl hypochlorite, -hypobromite, -nitrite, -chromate, -acetate; triphenylmethyl-hypochlorite, -chlorate, -bromate, -iodate, -nitrite, and *n*-butyl nitrite react by rupture of the oxygen bond in the anion; *t*-butyl nitrate, triphenylmethyl-perchlorate, and -nitrate by the rupture of the C—O bond; and *n*-butyl nitrate, *n*-octyl nitrate, ethyl sulfate, biphenylmethyl chromate, and -acetate by mixed mechanisms. The hydrolysis of a number of hydroperoxides of secondary and tertiary alcohols has been shown (117) to take place by C—O bond rupture. Khaskin (52) finds that in the hydrolysis of $Si(OC_2H_5)_4$, the Si—O bond breaks; but in the hydrolysis of $Si(OCOCH_3)_4$, the C—O bond breaks.

In the reaction of ΦSO_3Na with alkali to form ΦONa and sulfite, solvent oxygen appears in the phenol (118). This result is not surprising if it is admitted that there is a C—S bond in the sulfonate, but it does not establish the existence of such a bond, because exchange between the parent compound and H_2O was not measured. Bunton & Frei (8) found that in alkaline hydrolysis of phenyl toluene-p-sulfonate, the S—O rather than the Φ—O bond is ruptured. The Φ—O bond is apparently particularly resistant to substitution reactions; thus exchange of Φ—OH with water has not been observed even under the most drastic conditions (56). The reaction of $(C_2H_5)SO_4$ with C_2H_5OH in OH^- to form diethyl ether takes place by cleavage of the C_2H_5—O bond in the ester (119).

Reactions involving organic phosphates and biological derivatives.—There is especially great interest in the point of cleavage in phosphate derivatives, because of the importance of C—O—P, P—O—P and N—P bonds in metabolic processes. The experiments done in this field have included the study of enzymes as well as acids and bases as catalysts.

Blumenthal & Herbert (120) showed that on the hydrolysis of $(CH_3)_3PO_4$ in an acid solution, or one which is initially neutral, the C—O bond is broken, but that in alkali, the P—O bond breaks. Bentley (121) has shown that for acetyl phosphate, however, the P—O bond ruptures in acid. The conclusion that the C—O bond is broken in the alkaline hydrolysis is not so clear-cut. For both sets of conditions, the conclusion is based on isotopic analysis of the acetate and not of phosphate oxygen. Exchange of the ester (88), as well as C—O fission could cause solvent oxygen to appear in the acetate. With acetylphosphatase as catalyst (121), C—O cleavage occurs.

In the alkaline hydrolysis of ribonucleic acid, one solvent oxygen is placed on each phosphate formed (122). In the acid hydrolysis of glucose-1-phosphate the C—O bond is broken, but under the influence of acid and of alkaline phosphatases, the P—O bond breaks (16). This type of cleavage occurs also when alkaline phosphatase acts on adenosine 3-phosphate, adenosine 5-phosphate, β-glycerophosphate and phenyl phosphate (123). Stein & Koshland (123) point out that in view of the agreement of the

isotopic composition of the phosphate with that expected on placing one solvent oxygen on each ion, a 5-coordinated intermediate, analogous to the 4-coordinated intermediate proposed by Bender (88) for hydrolysis of some organic esters, cannot be involved. The hydrolysis of ATP catalyzed by lobster muscle strips (124) or by muscle protein (125) also produces $PO_4^{\equiv}$ with a single oxygen derived from the solvent.

In the formation of glucose-1-phosphate from glycogen under the influence of muscle phosphorylase, the P—O bond is preserved, and practically no change in isotopic composition occurs in the time required for the system substantially to reach chemical equilibrium (16).

Phosphate oxygen has been traced in several systems involving the ADP-ATP transformations coupled to other changes. Stulberg & Boyer (126) have found that in the reaction

$$\text{Citrulline} + PO_4^{\equiv} + \text{ADP} \rightarrow \text{ornithine} + NH_3 + CO_2 + \text{ATP}$$

phosphate oxygen appears in the CO_2 to the extent of *ca.* 28 per cent of that expected if CO_2 forms directly without passing through $CO_3^{=}$. The reaction:

$$\text{glutamate} + NH_4^+ + \text{ATP} \rightarrow \text{glutamide} + \text{ADP} + PO_4^{\equiv}$$

proceeds with one (127, 128) glutamate oxygen appearing in each $PO_4^{\equiv}$ and one (128) in ADP. The reaction:

$$\text{ATP} + \text{acetate} + \text{Coenzyme A} \rightarrow \text{adenylic acid} + \text{acetyl-Coenzyme A} + \text{pyrophosphate}$$

takes place with acetate oxygen appearing in the adenylic acid and essentially none in the pyrophosphate (127). Apparently in the glutamate system, nucleophilic attack on the terminal $PO_4^{\equiv}$ occurs; and in the acetate-coenzyme system in the phosphate attached to the ribose. Harrison, Boyer & Falcone (129) have shown that when 3-phosphoglycerate is formed by the enzymatic oxidation of glyceraldehyde-3-phosphate in the presence of inorganic phosphate, oxygen of the inorganic phosphate is found in the carboxyl group of the glycerate. Thus, in the intermediate stage involving transfer of phosphate from 1,3-diphosphoglycerate to ADP, cleavage occurs in the P—O bond of the COP linkage.

Some of the observations cited above show clearly that the enzyme rather than the composition of the medium determines the mode of attack in a particular reaction. The observations on the point of cleavage in the enzymatic hydrolyses of phosphates, and in the action of phosphorylases, provide part of the basis for the generalization by Koshland & Stein (130) that in the hydrolysis of ROR′ by an enzyme, the oxygen bond is split in the moiety for which the enzyme shows specificity. The generalization finds additional support in the observation (130) that under the influence of invertase, the fructose—O bond in sucrose is split, and in the hydrolysis of some glucosides, catalyzed by glucose-specific glucosidases, the glucose-O bond is ruptured (131).

Tracer work has also been done on phosphates as they are involved in oxidation-reduction reactions. Cohn (132) observed that in the oxidation of the substrates α-ketogluterate, β-hydroxybutyrate, and succinate in the

presence of rat liver mitochondria using adenylic acid as acceptor for inorganic phosphate, a rapid turnover of oxygen in the phosphate occurs. This exchange occurs only when phosphorylation takes place. It is suppressed by dinitrophenol and does not occur when Mg^{++} and adenylic acid, both necessary for phosphorylation, are absent.

Boyer, Falcone & Harrison (133) report that liver mitochondria catalyze the $PO_4^{\equiv} - H_2O$ exchange even when O_2 or substrate is absent. The oxygen exchange rate much exceeds that of the phosphate exchange with ATP as measured using P^{32}, and therefore the oxygen exchange cannot be attributed to the equilibrium:

$$\text{ADP} + \text{phosphate} \rightleftharpoons \text{ATP} + H_2O.$$

The work has been further extended by Cohn & Drysdale (15) using adenylic acid or glucose as phosphate acceptors, and making oxygen-isotopic analyses of both the inorganic phosphate and the bound phosphate. When labelled $PO_4^{\equiv}$ is used in ordinary water, the oxygen in the bound phosphate is observed to have undergone more exchange than corresponds to the average isotopic composition of the inorganic phosphate during the time of the experiments. Furthermore, using H_2O^{18}, but with a phosphate of ordinary isotopic composition, the extent of exchange of inorganic phosphate is unaffected, but that bound in ATP is reduced. These results show that more than one intermediate is involved between inorganic phosphate and bound phosphate, and furthermore, that some species, not readily exchangeable with water, furnishes oxygen to the phosphate which is eventually bound.

Reactions involving complex cations.—The problem of position of bond rupture is active also for oxy-anions bound to metal ions. Thus, when $Co(NH_3)_5CO_3^+$ reacts in acid to form $Co(NH_3)_5H_2O^{+++}$ and CO_2, the oxygen in the bound water does not come from the solvent, but is left by the carbonate (134); and CO_2 is formed without admixture of solvent oxygen (67, 135). In the reaction of $Co(NH_3)_4CO_3^+$ with acid, in which the product $Co(NH_3)_4(H_2O)_2^{+++}$ is formed, one oxygen in the aquo ion comes from the solvent, and one is left behind by the $CO_3^{=}$ when CO_2 is released (67). Presumably, the ion $CO(NH_3)_4H_2O\ CO_3^+$ is first formed, picking up oxygen from water, and in the final act, the mechanism is like that described for $Co(NH_3)_5\ CO_3^+$.

In the alkaline hydrolysis of the series of pentamminecobaltic complexes containing acetate, monochloracetate, trichloracetate, and trifluoracetate as sixth groups, the resulting organic ions were found (135) to contain solvent oxygen corresponding to the rupture of C—O to the extent of 4, 7, 20 and 53 per cent respectively for the four systems. Exchange of the complex ion with solvent was not studied, so that it is not possible to say with certainty that these results measure the extent of C—O bond breaking.

The hydrolysis of the ion $Co(NH_3)_5ONO^{++}$ in acid proceeds with O—N bond rupture, but in alkali with Co—O rupture (136). This change in mechanism is consistent with the general behavior of oxy-anions and of cations. For anions, lability to substitution increases with acidity; for

cations attached to ligands having dissociable hydrogens, it increases in more alkaline solutions. There is no oxygen exchange with solvent in the rearrangement of the nitrite ion, $Co(NH_3)_5ONO^{++}$ to the corresponding nitro form (136).

Other reactions.—The interesting result obtained by Wang & Tarr, that in the hydrolysis of urea by the action of urease, precisely one-half the oxygen in CO_2 is derived from the solvent (137), can be considered in the context of the results for carbonate complexes outlined in the preceding section.

Some of the conclusions reached by Bernstein & Pars (138) on the precipitation of metal oxides in alkaline solution are subject to modification (139) in the light of more recent work they have done which is still unpublished.

Oxidation-Reduction Reactions

An important application of oxygen isotopes is to learn the source of oxygen added to the reducing agent when it is acted on by an oxy-oxidizing agent in solution. In considering a solvent such as water, having labile H—O bonds, this problem is not formulated merely to give point to tracer experiments. Reactions are in fact known in which the oxygen taken up by the reducing agent does come from the water rather than from the oxidizing agent. This result would always be obtained if the reaction were to take place with oxidant and reductant separated, but can also be obtained if the reducing agent attacks the central atom of an oxy-anion, rather than one of the oxygens. The reactions of ΦH with H_2SO_4, which produces ΦSO_3H as an intermediate stage, and which on reaction in alkali forms ΦONa having oxygen derived from the solvent (118), is an example in the last category. Another is the reaction of nitrite and sulfite forming as final products NH_2OH and $SO_4^{=}$. The intermediate $HON(SO_3)_2^{=}$ can be isolated in this system, and when this is hydrolyzed solvent oxygen is placed on the $SO_4^{=}$ (140). These examples taken in the context of the first few in which the oxygen atom is shown to be transferred, will serve to show that the path of oxygen for reactions involving oxy-ions requires experimental investigation.

Oxidations by molecular oxygen.—Winter & Briscoe (41) found that in the oxidation of sulfite by O_2, one atom in the sulfate is derived from the oxygen. This result was confirmed in its essential feature by Halperin & Taube (141), although the work of the latter authors suggests that slightly more than one oxygen-O appears in each sulfite. Mason, Fowlks & Peterson (142) have found that in an enzymatic oxidation catalyzed by the phenolase complex, the atoms of O_2 do appear in the product phenol. Careful studies by Rubin *et al.* (143) have shown that the so-called additives in the photochemical production of H_2O_2 in water sensitized by ZnO actually serve as reducing agents for molecular O_2. This conclusion has been verified (144) by tracer experiments which have demonstrated that the oxygen in H_2O_2 is derived from O_2.

Oxidations by oxy-halogen compounds. Fairly extensive tracer studies have been made with halogen-oxygen species as oxidizing agents (14). With

$SO_3^=$ as reducing agent and ClO_3^-, ClO_2, ClO_2^-, or BrO_3^- as oxidizing agents, the oxygen of the oxidizing agent, except for a defect of *ca.* 0.5 in the case of the chlorine compounds and 0.8 in the case of BrO_3^-, is transferred completely to $SO_3^=$. These results in themselves, and more direct observations with Cl_2O, suggest that the defect occurs in the stage of ClO^- or BrO^- reduction. Thus for the stages $ClO_3^- \rightarrow ClO_2^- \rightarrow ClO^-$, transfer may be assumed complete, with the reducing agent attacking the oxidizing agent at an oxygen atom. With ClO^-, this mechanism operates in part, but presumably also an alternative one in which Cl^+ is in effect transferred from HOCl. The reaction of ClO^- with nitrite in acid takes place with essentially complete transfer of oxygen from ClO^- to NO_2^- (37).

Transfer is observed also in the oxidation of metal ions by ClO^- (17). Thus, when $Pb(NO_3)_{2aq}$ is added to moderately alkaline ClO^-, a partially oxidized product is formed at once, which contains ClO^--oxygen corresponding nearly to the oxidation as:

$$Pb^{++} + 2OH^- + ClO^{*-} \rightarrow PbOO^* + H_2O + Cl^-.$$

In strong alkali, starting with plumbite ion, reaction is still rapid, but precipitation of a product is long delayed. When a solid does finally form, it may contain no ClO^- oxygen. When $Pb(OH)_2$ is formed before ClO^- is added, the extent of oxidation exceeds the extent of transfer, and in this case, oxidation by electron transfer through the solid apparently takes place. Except for the features involving the amphoteric behavior of Pb^{++}, the observations with Mn, Co, and Ni salts are similar to those outlined. The oxygen liberated when ClO^- decomposes in the presence of Co, Cu, or Ni oxides comes essentially completely from water.

An isolated result which can best be mentioned in relation to the results just described on oxygen atom transfer in forming metal oxides is that obtained (141) on the oxidation of $SO_3^=$ to $SO_4^=$ by MnO_2. Each $SO_4^=$ formed is found to contain one oxygen atom derived from the metal oxide.

Oxidations by peroxides.—Reactions in which H_2O_2 is the oxidizing agent have revealed some interesting and unexpected features of mechanism in the course of oxygen tracer studies. The result (141) that in the reaction

$$H_2SO_3 + H_2O_2 \rightarrow H^+ + HSO_4^- + H_2O$$

two of the oxygen atoms in each sulfate originate in the peroxide, suggests a mechanism in which substitution of OH in H_2SO_3 by OOH first takes place, and the resulting permonosulfurous acid then rearranges to $SO_4^=$. This mechanism finds support in the observation that in alkaline solution only a single oxygen atom is transferred for each sulfate formed. (17) In alkaline solution, O in $SO_3^=$ is not readily replaced by OOH, and the system finds direct transfer of oxygen from peroxide to intact $SO_3^=$ a more ready access to products.

In the reaction of H_2O_2 with NO_2^-, a pernitrite has actually (145) been demonstrated as an intermediate. Tracer experiments on this system (78) have shown that at low NO_2^- concentrations, in analogy to the sulfite reac-

tion, H_2O_2 places two of its oxygen atoms in each NO_3^- formed. When the nitrite concentration is high, the extent of transfer decreases, but not to as little as one oxygen per H_2O_2 reacting. During the reaction H_2O_2-oxygen appears in NO_2^-, although NO_2^--oxygen does not appear in H_2O_2. These results find a reasonable explanation in the following steps:

$$HONO + H_2O_2^* \rightarrow HO^*O^*NO + H_2O$$

$$HO^*O^*NO \rightarrow ONO_2^{*-} + H^+$$

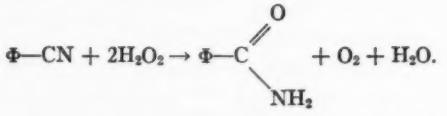
$$HO^*O^*NO + NO_2^- \rightarrow O^*NO^- + H^+ + O^*NO_2^-.$$

Although direct attack of H_2O_2 by NO_2^- is slow, the O—OH bond is sufficiently polarized in replacing H^+ by NO^+ to make transfer of O from HOONO to NO_2^- possible. The activation of the O—OH bond by electron-attracting groups is suggested also by the rapid reaction of NO_2^- with

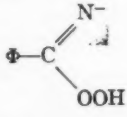
in which approximately only one oxygen atom is transferred (78) for each NO_3^- formed.

In the reaction of Pb(II) in alkaline solution with H_2O_2, substantially all the oxygen in the PbO_2 originates in the H_2O_2. Large extents of transfer can be observed also in the oxidation of Mn(II) and Co(II) by H_2O_2 (17).

A remarkable reaction is the hydration of ΦCN to an amide under the influence of H_2O_2 in alkaline solution:

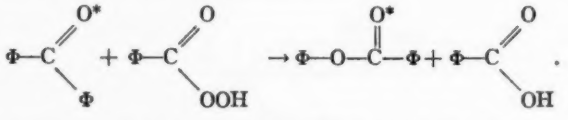


Wiberg (146) has shown that the oxygen in the amide is derived from H_2O_2 rather than from water. He suggests the species

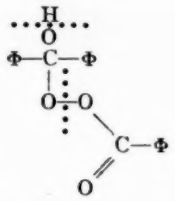


as an intermediate, this reacting by oxidizing another molecule of H_2O_2.

The isotope-course and stoichiometry of a peroxide reaction studied by Doering & Dorfman (9) can be represented as:



The tracer experiment eliminates the Wittig and the Baeyer mechanisms, but is consistent with that proposed by Criegee:



Oxidations by permanganate.—In the oxidation of sulfite by MnO_4^- in alkali, very little MnO_4^- oxygen appears in the sulfate (141). Very little oxygen transfer is observed (147) in the alkaline oxidation by MnO_4^- of an aromatic aldehyde, although in neutral solution, the reaction does take place by oxygen transfer. The mechanisms of the reactions not involving transfer are unclear. They may take place by a complex process in which MnO_4^- is first reduced by a lower oxidation state of Mn, and the reducing agent is then attacked by the resulting intermediate, which undergoes exchange before it reacts.

It has also been suggested (148, 149) that MnO_4^- can oxidize HO^- sufficiently rapidly (forming HO or O^-), so that this process can carry the reaction; i.e., O^- (or HO) rather than oxidizing agent attacks the reducing agent. Symons (149) finds in this process a plausible explanation of his important tracer results which showed that the oxygen which is formed when MnO_4^- decomposes in aqueous alkaline solution comes from the water, and that the $MnO_4^=$ formed retains the isotopic composition of the MnO_4^-. However, the reaction:

$$MnO_4^- + HO^- \rightarrow HO + MnO_4^=$$

is subject to the free energy requirement $\Delta F^\circ \sim 34$ kcal mole^{-1} (150). Even making allowance for the possibility that O^- rather than HO may be present in alkaline solution:

$$MnO_4^- + 2HO^- = MnO_4^= + O^- + H_2O,$$

and using for the acid dissociation constant of HO a value of 10^{-10} [which appears reasonable in comparison to the known (150) value of 10^{-12} for that of H_2O_2, and with certain experimental results (151)], ΔF° is strongly positive, $\sim$29 kcal mole^{-1}.

It seems unlikely that a path as energetically unfavorable as this can provide the path for a rapid oxidation. The question can be raised whether the decomposition of MnO_4^- as ordinarily observed is in fact a homogeneous reaction. For a path involving a solid such as MnO_2, it is no longer proper to describe the reactions by equations of the kind which have been written above.

The mechanism of the photochemical decomposition of MnO_4^- in water is in marked contrast to that described for the spontaneous reaction. Zimmerman (152) has shown that in the photochemical reaction, the oxygen formed comes entirely from the ion.

Decomposition and oxidation of hydrogen peroxide.—Tracer results

(40, 153, 154, 155) on the decomposition of H_2O_2 in solution are in marked contrast to those described for the spontaneous decomposition of MnO_4^- and for the catalyzed decomposition of ClO^-. Under all conditions tested, the oxygen liberated originates entirely in the water. These include: the spontaneous decomposition; the decomposition catalyzed in homogeneous solution (e.g., by Fe^{++}—Fe^{+++}, I^-—I_2, Br^-—Br_2, Cr(VI)—Cr(?), catalase); the decomposition on surfaces (e.g., of various metals, MnO_2, PbO_2, Co_2O_3, MnO_2); and the photochemical decomposition (156). A question which is related; since the liberation of oxygen in the catalytic decomposition presumably involves the action of an oxidizing agent derived from the catalyst—is that of the source of oxygen produced when hydrogen peroxide acts as a reducing agent. For all oxidizing agents which have been tested (Ce(IV), Cl_2, ClO^-, MnO_4^-, $CrO_4^=$) the O_2 originates entirely in the H_2O_2. The difference between ClO^- and H_2O_2 noted in the tracer experiments on the decompositions is in agreement with other well-known differences in their behavior. ClO^- is very difficult to oxidize, and thus H_2O rather than ClO^- acts on the oxidizing agent characteristic of the catalyst; whereas H_2O_2 is known to be a much more rapid reducing agent than is water.

Reactions involving hydroxyl radical and oxyl ion.—There are a number of observations which relate to the important problem of the reactions of hydroxyl radicals in water. While there is doubt that MnO_4^- can cause a rapid oxidation of HO^- to HO, there is little doubt that the radical-ion SO_4^- is energetically capable of bringing this about. Evidence in support of such a reaction is derived from work done by Kolthoff & Miller (157). In the acid decomposition of $S_2O_8^=$, the oxygen comes solely from the ion (hydrolysis to H_2O_2 presumably is a first step); in alkaline solution the oxygen comes only from the solvent. It is likely (157) that the first step in the decomposition in alkali is rupture of the O—O bond, forming SO_4^-, and that SO_4^- then oxidizes HO^-.

Another interesting feature of the reaction is that in alkaline solution, but not in acid, the decomposition of $S_2O_8^=$ induces the exchange of O_2 with water (158). Turnover numbers as high as 200 may be attained, but the reaction is not readily controlled as it appears to be inhibited by impurities leached from the glass reaction vessel. Hart and coworkers (99, 151) have reported that the same exchange reaction is induced by X-rays or ultraviolet light. These workers noted further that the effect is greatly diminished by the presence of H_2O_2. A similar exchange effect takes place also in the alkaline decomposition of O_3 (39). The isotopic composition of the O_2 formed shows that much oxygen in addition to that produced by the decomposition of O_3 reaches isotopic equilibrium with water. The excess exchange reaches a maximum at moderate alkalinity, and disappears in acid. These induced exchange effects may well have a common explanation. They may involve the oxyl ion O^- as an intermediate which can exchange with O_2 and with HO^- (99).

When O_3 decomposes in acid solution, the isotopic composition of the oxygen shows that there it has undergone a slight exchange with water. This

exchange is inhibited by certain agents, among them HOAc, Cl^-, and NO_3^- (at high concentration). When the concentration of O_3 becomes very low, the exchange approaches one-sixth of the O_3 which has decomposed. These observations constitute strong evidence that the radical HO is in fact involved as an intermediate in the decomposition. On the basis of the following reaction sequence involving HO, the limit of one-sixth exchange is easily understood: (39)

$$HO + H_2O^* \rightarrow HO^* + H_2O$$

$$HO^* + O_3 \rightarrow HO^*O + O_2$$

$$\underline{HO^*O + O_3 \rightarrow HO + OO^* + O_2}$$

$$2O_3 + H_2O^* \rightarrow 2O_2 + OO^* + H_2O$$

Reactions of oxy-nitrogen compounds.—Bothner-By & Friedman (159) have combined nitrogen and oxygen tracer work in a study of the reaction of hydroxylamine with nitrite, which yields N_2O as product. Assuming the nitrite, but not the hydroxylamine, to be enriched in N^{15}, the results can be described as follows. In alkaline solution, $N^{15}NO$ and $NN^{15}O$ are produced in equal amounts; and when the reaction is carried out in O^{18}-enriched water, none of the solvent oxygen appears in the product. In acid solution, there is a preponderance of $NN^{15}O$; and O^{18} appears in combination $NN^{15}O^{18}$ but not as $N^{15}NO^{18}$. The results show that a symmetrical intermediate is produced when the reaction takes place in the less acidic solution, and that a path involving an unsymmetrical intermediate comes into play when the solutions are acidic. The oxygen isotope results can be understood in terms of an acid-labilized exchange of oxygen between NO_2^- or the unsymmetrical intermediate and solvent, with NH_2OH resisting exchange.

Tracer studies have been done on the reaction (160):

$$2NO_2 + 2OH^- = NO_2^- + NO_3^- + H_2O.$$

Only under extreme conditions (very low partial pressure of NO_2 in the carrier gas) is a tracer result obtained in harmony with the view that N_2O_4 submits to reaction in the unsymmetrical configuration:

$$ONONO_2 + 2O^*H^- \rightarrow ONO^{*-} + NO_3^- + H_2O^*.$$

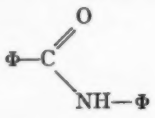
Under most conditions, each NO_3^- contains approximately one oxygen derived from the solvent, while the isotopic composition of NO_2^- varies from experiment to experiment.

Photosynthetic production of molecular oxygen.—Experiments have been done (161, 162, 163) to distinguish between $CO_3^=$ and H_2O as the immediate source of O_2 in photosynthesis, and the observation has been reported that the O_2 has the isotopic composition of the water rather than of the $CO_3^=$. In the absence of experiments testing the rate of exchange between $CO_3^=$ and H_2O in functioning cells, these results cannot be taken as proof of the conclusion that H_2O rather than $CO_3^=$ is attacked in liberating O_2. In connection with these studies, it is interesting to note that there is no O_2—H_2O exchange during photosynthesis, nor is there O_2—H_2O exchange during respiration

(161). An experiment to learn whether solvent oxygen is taken up by carotenoids during photosynthesis gave inconclusive results, because of difficulty in analysis (164).

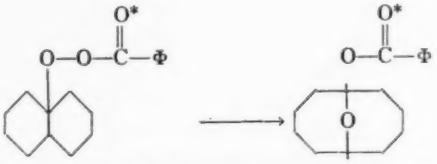
Molecular rearrangements involving oxygen migration.—Some reactions in this category were discussed in the section on oxidation reactions of peroxides.

In the Beckmann rearrangement of Φ_2CNOH, the oxygen in the product



is found to have undergone isotopic exchange with the water (165, 166). This observation is consistent with the interpretation that the reactant oxygen is lost during reaction, but requires a study of the rate of exchange of the parent compound with water, to strengthen this interpretation to the quality of a conclusion.

An interesting tracer result obtained by Denney (167) is described by the equation:



The carbonyl oxygen remains distinct from the others, thus proving that at no stage are the oxygens in ΦCO_2 allowed to become equivalent.

Fractionation Effects

Fractionation effects do not appear to have been studied and exploited as fully in purely chemical systems as they have in geochemical ones (1, 2), but several of the studies completed illustrate the use of fractionation data in advancing the understanding of mechanisms of reactions. Several equilibrium fractionation effects have been referred to already, and this subject will not be discussed further, except to cite papers by Urey & Greiff (168) and by Urey (169) as sources of data useful in calculating the equilibrium constants of a variety of reactions involving O^{18} exchange.

Reactions of hydrogen peroxide.—There has been some interest in kinetic fractionation effects in reactions of H_2O_2 (154, 155). The fractionation factor, f, for which values will be cited hereinafter is defined as follows:

$$= \frac{d\ ln\ (O^{16})}{d\ ln\ (O^{18})}$$

where (O^{18}) and (O^{16}) are the concentrations or amounts of the respective unreacted isotopes. For the reduction of H_2O_2 by Fe^{++}, Sn^{++}, Cu^{+}, Cr^{++}, and Ti^{+++}, f has values 1.068, 1.06, 1.055, 1.050 and 1.006, respectively. The similarity in the magnitude of the fractionation effects for Fe^{++}, Sn^{++}, Cu^{+} and Cr^{++} suggests a common mechanism for the reaction of these substances

with H_2O_2, which is different from that realized with Ti^{+++} as reactant. It may be significant in this connection that Ti^{+++} is the only one of the reducing agents which is restricted to a one-electron oxidation process. The fractionation effects observed for oxidizing agents attacking H_2O_2 are considerably smaller (1.011 for Ce(IV) and 1.009 for Cl_2) than for most reducing agents, and in view of the circumstance that the O—O bond is severed in reduction, but an O—H bond in oxidation, this is not an unexpected result. The fractionation effects of homogeneous catalysts for the decomposition can be resolved (154) into a factor f_r, corresponding to reduction of H_2O_2 in the catalytic cycle, and a factor f_o, corresponding to the complementary process, the oxidation of H_2O_2. The change in isotopic composition of the H_2O_2 is governed by the factor $(f_o+f_r)/2$. One can obtain f_o directly by comparing the isotopic composition of O_2 formed intially with that of the H_2O_2. The factors f_o and f_r are properties of the intermediates responsible for the reactions in question and can be used to identify them. Thus, the factor f_r measured for H_2O_2 being decomposed catalytically by Fe^{+++} is 1.068 (154), and the agreement with the value of f measured for Fe^{++} acting on H_2O_2 is evidence that Fe^{++} is in fact the reducing intermediate in the reaction.

There is a substantial isotope fractionation effect in the photochemical decomposition of H_2O_2, even for conditions under which the chain processes have been eliminated (156). In acid solution, $f_o=1.010$ and $f_r=1.005$; when acid is absent, f_o is essentially unchanged and f_r becomes 1.023. The marked change in f_r with acidity is not understood. It should be pointed out that the authors (156) failed to appreciate that there can be an isotope fractionation effect because of differences in extinction coefficients for the isotopically distinct molecules (170).

Aquation of complex cations.—The principle of using isotopic fractionation data to characterize intermediates has been applied also to the problem of mechanism of aquation of complex ions (171). If a common intermediate is formed in a series of reactions such as

$$Co(NH_3)_5Cl^{++} + Hg^{++} + H_2O \rightarrow Co(NH_3)_5H_2O^{+++} + Hg\,Cl^{+}$$

$$Co(NH_3)_5Cl^{++} + Tl^{+++} + H_2O \rightarrow Co(NH_3)_5H_2O^{+++} + TlCl^{++}$$

$$Co(NH_3)_5I^{++} + Hg^{++} + H_2O \rightarrow Co(NH_3)_5H_2O^{+++} + HgI^{+}$$

the isotopic fractionation in the formation of the aquo ion should be the same for all. If S_N2 mechanisms operate, the activated complexes contain, in addition to H_2O, the metal ion and halide ion and therefore differences in isotope fractionation can be expected for the different reactions. The summary of the observed f values is as follows:

	Hg^{++}	Ag^{+}	Tl^{+++}
$Co(NH_3)_5Cl^{++}$	1.012	1.009	0.996
$Co(NH_3)_5Br^{++}$	1.012	1.007	0.993
$Co(NH_3)_5I^{++}$	1.012	1.010	1.003

The results suggest that there are important contributions to the mecha-

isms by S_N2 paths for Tl^{+++} and Ag^+, as reactants, and suggest essentially pure S_N1 mechanisms with Hg^{++} as reactant.

Decomposition of ammonium nitrate.—Oxygen isotope fractionation has been observed in the thermal decomposition of NH_4NO_3 to N_2O and H_2O (48). On complete decomposition, the ratio O^{18}/O^{16} in N_2O compared to that in product water is 1.023 ± 0.003. Since the formation of water requires breaking of N—O bonds and the formation of N_2O does not, an effect in this direction is to be expected.

Photosynthetic production of molecular oxygen.—The fractionation of O_2 produced in photosynthesis (172) has been determined as 1.003, and this factor is regarded as being too small to account for the Dole effect (173).

Oxidation of metals.—There is considerable fractionation of the oxygen isotopes during oxidation of metals. Dole & Lane (174) reported the fractionation factors in the chemisorption reaction with Fe as 1.025–1.028, and in the chemisorption reaction with Cu as 1.052 to 1.074, at 25°C. Bernstein (175) has made a thorough investigation of the effects for oxide films of intermediate thickness (150–2500 A°) on copper. The factor is independent of film thickness over the range studied, is independent of pressure, and decreases with increasing temperature. When extrapolated to 25°C., an upper limit of 1.029 is found, significantly lower than the value reported (174) for very thin films.

Electrolytic production of molecular oxygen.—The fractionation of oxygen isotopes in electrolysis has also been a subject of considerable interest, although little systematic work on this problem has been done. Much of the earlier work is reviewed in a paper by Tronstad & Brun (176). More recently, some measurements have been made (177) comparing directly the isotopic composition of the oxygen formed electrolytically to that of the oxygen which is formed when water is oxidized completely to O_2 by BrF_3. The fractionation factors varied from 1.004 for Sn to 1.018 for Fe. For a given metal, subjected to fixed treatment, they are reproducible, and are independent of the current density at least over a range. The factors are sensitive to the nature of the surface, and in some cases to the presence of certain substances in the solution.

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RECENT ADVANCES IN LOW LEVEL COUNTING TECHNIQUES[1]

By Ernest C. Anderson and F. Newton Hayes

Biomedical Research Group, Los Alamos Scientific Laboratory, University of California, Los Alamos, New Mexico

Introduction

Problems involving low level counting techniques are encountered in many diverse fields including biological and chemical tracer studies, archaeological and geological dating (1), investigations of natural and induced radioactivities, and the study of fundamental particles. Such a host of methods and variations on methods have been reported that an exhaustive catalog would constitute little more than an annotated bibliography. We have, therefore, limited our attention to a few selected examples which seem to be of special interest because of their versatility or their extreme sensitivity. We prefer to organize our discussion around specific problems rather than around types of detectors since this permits a comparison of alternative methods and provides practical examples of various techniques in actual operation. Alpha counting is arbitrarily excluded. For a discussion of photographic methods (radioautographs) the reader is referred to Norris & Woodruff (2) and references cited therein. We will discuss the methods developed for the measurement of natural radiocarbon and tritium, cosmic ray produced Be^7, and the K^{40} content of the human body. Double-beta decay experiments are considered since they seem to constitute one of the most difficult of the outstanding problems and because of their importance from the viewpoint of theoretical nuclear physics. Finally, an account is given of recent attempts to detect the neutrino, probably the most difficult low level counting problem of all and one which seems finally on the verge of successful solution.

Principles.—The fundamental principles of low level counting are simple: maximize the number of detected events of the desired kind (signal) and minimize all others (background). The number of detected events is, of course, the product of sample size, over-all counting efficiency, and counting time. However, the striking importance of the net sample counting rate is not always fully appreciated. It can be shown (3) from elementary counting statistics, that if the sample rate is small compared with background (which is usually the case in a truly low level problem) and if counting statistics is the limiting source of error (also usually true), then the proper parameter for the comparison of counting methods is S^2/B, the ratio of the square of the net sample rate to the background rate. (Any sample strength can be used in making the comparison so long as the same strength is used in testing all methods.) The method giving the maximum value of S^2/B will yield the

[1] The survey of literature pertaining to this review was completed December, 1955.

greatest statistical accuracy in a given counting time. (If the sample rate is not small compared with background, then S is even more heavily weighted.) If S/B were constant while S increased a factor of 10, S^2/B would increase by 10, and a given counting precision would be attainable in one-tenth the counting time. Since stability is often the limiting factor in low level work, a reduction in the time over which a given stability must be maintained is often an important gain. The factors preventing unlimited exploitation of the advantage of large samples are often limited availability of sample, cost and complexity of large counters, and failure of counting statistics to be the limiting factor. However, some of the examples discussed below will indicate that the process of increasing sample size can be carried much farther than is ordinarily done.

Minimizing extraneous background events is an equally important part of successful low level detection. The classical methods of background reduction are well discussed by Libby (4) including the comparison of lead, iron, and mercury as gamma shields and the use of anticoincidence Geiger counters to eliminate penetrating cosmic ray events of high specific ionization (principally μ mesons). With Geiger counters, massive shielding and anticoincidence circuitry are the only available methods of background reduction. With counters giving pulses proportional to the energy of the initiating event, energy discrimination is also possible. This is more efficacious with monoenergetic signals such as conversion electrons and gamma ray line spectra than it is for continuous beta spectra. For large detectors in which mesons can deposit large amounts of energy, energy discrimination can accomplish the same result as anticoincidence shielding. Because of the varying energy sensitivities of different detectors, it is difficult to predict the background of one from that of another. The relative importance of the various components of background will vary with the detector.

The problem of purity of shielding materials and of materials of construction is a serious one. The early experiments reported by Rutherford, Chadwick & Ellis (5) are very revealing in their implications of the universal distribution of natural radioactivities. The fallout of fission products from atomic weapons tests and the reuse of materials which have become slightly contaminated at reactor installations are increasing the problem. Miller *et al.* have recently shown that an important part of the background of NaI crystals in a low level installation is due to natural K^{40} and fission product Cs^{137} (6). The Subcommittee on Radiochemistry of the National Research Council Committee on Nuclear Science (Chairman, J. R. Arnold, Department of Chemistry, Princeton University) is actively concerned with the problem. Plans are under way to investigate the contamination levels in various materials of shielding and construction and to consider the possibility of stockpiling some important "clean" materials. The facilities of the Los Alamos Human Counter (see below) with a gamma ray sensitivity of 5×10^{-10}curie and a sample capacity of 150 kg. will be available for measurement of materials of interest to this program.

Beta Counting

The radio-nuclides produced by the cosmic radiation have provided a rewarding challenge for the development of low level counting techniques. Because of the possible applications of these techniques to large-scale industrial tracer studies, in addition to their well-known uses in archaeology, geology, and meteorology, they have considerable general interest.

Carbon-14.—The production and distribution of natural radiocarbon have been described in detail (4, 7) and a bibliography on natural radiocarbon has recently been published by Levi (8).

While the discovery of natural radiocarbon was made with two liter Geiger counters filled with isotopically-enriched methane, the original studies on the natural distribution and a considerable fraction of the published age determinations were performed with anticoincidence shielded screen-wall counters. These are described in detail by Libby (4). For natural radiocarbon studies the screen-wall counter seems to be inferior to the newer gas proportional counting techniques (see below), but some of its fundamental advantages for low level research are worth restating. Prominent among these is the great versatility of sample type. Since the screen-wall counter is used as a Geiger counter with conventional filling gases, it is a particularly easy counter to operate and the simplest of electronic circuits are needed for its use. The ease of interchanging sample and background makes for great stability and permits the measurement of small fractions of background especially when the double counter tube (9) is used. Finally, Geiger action permits the counting of extremely low energy particles.

A theoretical comparison of the screen-wall with the gas counter as a function of counter size has been given by Anderson & Levi (9). Their conclusion was that there is little choice in terms of sensitivity for counters of reasonable size. The successful routine use of C_2H_2 (10, 11), CO_2 (12, 13, 14) and CH_4 (15) at pressures above 1 atmosphere has shifted the balance in favor of the gas counter for C^{14} dating. The most important factor, however, is the comparative freedom from contamination enjoyed by the gas sample counter. Since almost all the chemical processing can be done in a closed vacuum system, the sample can be completely protected from possible contamination. As a number of laboratories using the screen-wall technique experienced severe contamination difficulties during periods of atomic weapons testing, this is a fundamental advantage. Proportional counting seems to be in greater favor than gas-sample Geiger counting because the lower voltage required and the better plateaus obtained at high pressures.

Of the three gases used, acetylene has the advantage of getting more carbon into the counter at a given pressure, the disadvantage of a reputation for explosiveness, particularly at high pressures (no accidents have occurred during its use, however). CO_2 is by far the simplest to prepare chemically, since the first step in any of the procedures is the preparation of CO_2. This advantage is partly offset by the extreme purity required for proper counting characteristics, but the purification problems have been mastered in several

laboratories. The advocates of CH_4 claim the conversion of CO_2 to CH_4 to be easier than the purification of CO_2. CH_4 is, of course, one the of the best of counter gases. Several comparisons of the ultimate sensitivities of the various methods have been published (16, 17, 18) and disagreement in the numbers in some of these tables results from different assumptions as to statistical limits and relative frequency of background determinations. Roughly, the original screen-wall method has a range of 25,000 yr., the acetylene method extends to 30 to 35,000 (pressure limited), while the CO_2 and CH_4 methods extend to 25 to 40,000 yr., depending on counter size and pressure.

The method capable in principle of the greatest sensitivity and accuracy is the liquid scintillation technique (16, 19, 20). Its inherent advantage over the gas counting method is that the sample is in a condensed phase with a density 1000 times greater than the gases and thus subtends a much smaller angle to external background. The result is the rather surprising one that any ordinary C^{14} liquid scintillation counter will detect natural radiocarbon so effectively that certain otherwise excellent solvents (e.g., *p*-cymene) cannot be used for ordinary counting because of the high (600 c./min./100 ml.) natural C^{14} background (21). Background reduction is not difficult and ordinary Pb shielding (2-in) suffices. Anticoincidence shielding is not necessary since the mesons give such large pulses in the liquid scintillator that they are easily eliminated with an upper gate. However, the use of liquid scintillation counting is greatly hampered by the complexity of sample preparation methods. One technique has avoided this difficulty by selecting as samples natural materials which are themselves good scintillation solvents (e.g., pinene from turpentine) and which, therefore, require no chemical treatment at all (16). The limitation in choice of samples resulting from these requirements appears to eliminate this method from consideration in dating problems. It does have important applications to the study of the contemporary assay.

The scintillation methods which aim at the counting of all types of samples attempt to synthesize a suitable scintillation solvent from the CO_2 from the sample. The difficulty of a total synthesis of a good aromatic solvent is such that alternative approaches have been adopted, one of which is the partial synthesis of a solvent such as toluene in which only the methyl side chain contains the sample (17, 22). The other approach is the synthesis of a suitable diluent, that is a liquid which is not a good scintillation solvent but which can be added to a scintillation solution in large amounts without seriously interfering with the light emission processes. Aliphatic hydrocarbons (23) and methyl borate (22) are good examples of this type. The solution of acetylene in a scintillator solution (24) has been studied but it does not appear possible to count a large enough sample to make full use of the possibilities of liquid scintillators in this way.

Scintillation methods are still too complicated in their chemistry to compete with gas counting for ordinary samples. Their potential usefulness is for very old samples or for very high precision measurements of recent samples. The range of these methods approaches 50,000 yr. for 48 hr. of counting time.

At this level, the specific activity of the carbon is about 0.03 min.$^{-1}$ gm.$^{-1}$.

A precision of 0.3% of the contemporary assay has been demonstrated by Arnold for 48-hr. counts with a liquid scintillation method (23). [A similar precision was obtained by Suess using acetylene gas counting and counting periods of several weeks per sample (25).] The use of "balance point" operation is an important part of the instrumentation method. By the proper choice of discriminator settings it is possible to operate a liquid scintillation counter so that C^{14} counting rate and background rate pass simultaneously through a maximum and a minimum value respectively as high voltage or amplifier gain is varied. At this point the slope of counting rate versus gain is zero for both sample and background and the effect of electronic drift is minimized. With this sort of precision, the fine structure of C^{14} mixing phenomena, such as the Suess effect (23, 25, 26), can be readily studied.

A very interesting application of natural radiocarbon techniques is the study of the sources of smog (27). These results indicate the feasibility of large scale industrial tagging at levels low enough to avoid any possibility of health hazards.

Tritium.—Natural tritium (28, 29) constitutes an even more difficult detection problem than natural radiocarbon because of the lower specific activity and the softer beta ray. Isotopic enrichment by a factor of the order of 10^3 has been the common practice before counting. The gas sample counter is the method of choice. Because of the extremely low energy of the beta rays, external sample methods are not sensitive. The diffusion cloud chamber (30) has been used but this method is laborious and subjective. The concentration of tritium in surface waters (rain, snow, rivers) is approximately 10^{-17} (atom fraction in hydrogen) so that the specific activity is of the order of 0.2 min.$^{-1}$ (gm. H)$^{-1}$.

Libby's method of natural tritium measurement (31, 32, 33) uses water samples of 10 to 100 l. volume. These are distilled and then electrolyzed as a NaOH solution with iron-nickel electrodes to a residual volume of 1 cc. or less. The tritium enrichment is calculated from the measured deuterium enrichment. The enriched water sample is converted to H_2 gas by reduction with Zn dust at 500°C. It is then counted in a one liter Geiger counter using a filling of 2 cm. ethylene, 3 cm. argon, and up to 30 cm. of hydrogen. The counter is installed in an 8 in. iron shield with anticoincidence shielding. Background is about 8 c./min. and the tritium samples usually counted 5 to 10 c./min. above background. Counting time is half an hour.

Applications of natural tritium measurements (34) and industrial applications of low level methods (35) have been discussed by Libby.

Measurements of the tritium content of atmospheric hydrogen have been reported by Faltings & Harteck (36) and by Grosse *et al.* (37).

Gamma Counting

Beryllium-7.—Be^7 decays by electron capture with a 53 day half-life and the emission of a 478-Kev. gamma ray. Arnold & Al-Salih (38) have demon-

strated the existence of natural Be^7 by precipitating $Be(OH)_2$ carrier from 5–50 gal. samples of rainwater and counting the purified precipitate with a 1-in. NaI(Tl) crystal.

Although this cosmic-ray produced nuclide has a much lower production rate than either C^{14} or H^3, it is considerably easier to detect. This is due partly to the fact that it is not diluted in nature with large amounts of the stable isotope and partly to the greater ease of detection of gamma radiation. The sharp photo-peak obtained from monoenergetic gamma radiation with a NaI scintillation crystal spectrometer makes it possible to reduce background to low values by counting in a narrow energy band centered on the peak. Measurements were made with a low level spectrometer developed for the study of the decay scheme of natural Lu^{176} (39, 40).

Be^7 should be useful for the study of mixing in the atmosphere.

Potassium-40.—The possibilities of low level gamma spectrometry have been highly developed by Marinelli and his co-workers in their studies of the radioactivity of the human body (41, 42). The activities normally present are C^{14} and K^{40}, with H^3 and members of the U-Ra series at a much lower level (43).

The potassium of the human body amounts to about 150 gm. in the average adult (44). Since the gamma specific activity is 3 $sec.^{-1}$ $gm.^{-1}$, this amount corresponds to 450 d./sec. (about 0.01 μc.) in 70 kg., or 0.4 $min.^{-1}$ $gm.^{-1}$. This can be compared with the C^{14} specific activity of the human body of 2.7 $min.^{-1}$ $gm.^{-1}$. The measurement of the normal body potassium by external counting is, therefore, well within the realm of low level counting. Because of the comparatively low self-absorption of the body for gamma-rays of this energy (1.5 Mev), virtually the entire disintegration rate is detectable externally, the only limitation being that of counting geometry. Using a 4 in.$\times 1\frac{1}{2}$ in. crystal placed close the subject's back the net counting rate observed in the K^{40} photo-peak is about 11 c./min. from a typical subject against a background of 9 c./min. A counting time of one hour is, therefore, required to give a statistical error of 5 per cent. By using a larger crystal, counting time can be considerably reduced. The counter is contained in a special 8 ft.$\times$8 ft.$\times$6 ft. room with 8 in. thick steel walls (52 tons). Because of the ingenious construction of the room, four men can disassemble it for moving in two days. Materials of construction must be very carefully chosen to avoid contamination by Ra, K^{40} and fission products. A detailed analysis of the sources of background has been given by Miller *et al.* (6).

An alternative approach has been adopted at Los Alamos. Since the low rates and long counting times of the NaI crystal counter are due to the low geometrical efficiency, an increase in counting rate of several orders of magnitude can be obtained with a 4π well-type counter. The very large liquid scintillation detectors (45, 46) built for the neutrino program established the feasibility of a gamma counter large enough to contain a human subject (47). Such a counter has been built by the Biomedical Research Group of the Los Alamos Scientific Laboratory and is now in operation (48, 49). While its

background is astronomical by ordinary standards (about 400 c./sec. or 24,000 c./min.), the background is extremely stable (0.2 per cent over periods of hours) and the counting efficiency is so high (10–30 per cent for gamma rays above 0.5 Mev) that very small amounts of gamma emitters can be detected. For a counting time of 100 sec., the statistical precision is 2 c./sec. or about 5×10^{-10} curie. For a 100 kg. sample, this corresponds to a gamma specific activity of 0.01 min.$^{-1}$ gm.$^{-1}$. The natural K^{40} of man can be determined to a precision of 5 per cent in 16 sec. of counting time.

The counter consists of a cylindrical steel tank 30 in. in diameter and 72 in. long with an 18 in. diameter cavity running through the tank. The cavity has a thin (0.019 in.) stainless steel wall. The subject or sample is placed in the cavity as in a conventional "well-type" counter. The scintillation liquid [140 gal. of terphenyl plus POPOP in toluene (50)] surrounds the cavity in a layer 5 in. thick. Geometry is nearly 4π except for end loses (5 per cent for a point source in the center; average geometrical efficiency 90 per cent over the central 4 ft.). One hundred and eight 2 in. photomultiplier tubes (DuMont 6292) placed in the outer wall of the tank detect the scintillations. Conventional electronic circuitry is used.

The large liquid scintillation counter has considerably poorer energy resolution than the NaI crystal and it is not feasible to discriminate between two gamma rays unless their energies differ by a factor of two. However, its very short counting times should be of considerable value when large numbers of samples must be analyzed. It should be a useful instrument for measuring low level contamination of foodstuffs, water supplies, materials of construction and shielding, and personnel.

Mention should also be made of the measurement of human potassium by the ion chamber technique developed by Sievert in Sweden (51) and by Burch & Spiers in England (52, 53). While this perhaps represents a *tour de force* in pushing a classical detector to extreme limits, it illustrates the extent to which the inherent stability of ion chamber methods can be utilized by extremely careful and skilled techniques. The ion chamber is, of course, unable to distinguish events of the desired energy from background and anticoincidence shielding cannot be used. As a result, signal is a small fraction of background and very long integrating times are required. Large corrections are necessary for the reduction of background by the subject.

Double-Beta Decay

One of the major unsolved problems in radioactivity is the question of the existence of double beta-decay. This is the process in which the atomic number of a nuclide is changed by two charge units through the simultaneous emission of two beta particles. Many cases are known in which such a transformation is energetically possible, but the expected half-lives are extraordinarily long. The matter is of considerable theoretical importance because of the light it would shed on the nature of the neutrino (54). Stated very briefly, if the neutrino emitted in positron decay is different from that

emitted in negatron decay (the two neutrinos being a Dirac pair in the same sense as the positive and negative electrons are) then two neutrinos must be emitted in double beta-decay and the total emission of four particles will make the half-lives 10^{18} to 10^{19} yr. for 4 Mev decay energy (55). If, however, the β^- and β^+ neutrino are identical (Majorana case), then double beta-decay can occur with the virtual emission of a neutrino, the reabsorption of the same neutrino, and the net emission of only two particles, The half-life, for favorable cases, becomes as short as 10^{15} yr. for 4 Mev decay energy. These half-lives should be measurable with the best low level methods. Because of the omnipresence of trace amounts of ordinary activity and because of the complicated processes produced in counters by the cosmic radiation, the task of identifying double beta-decays is extremely difficult. It is rendered even less inviting by the fact that the odds are somewhat more than even in favor of the longer half-life so that there is a good probability of a negative result with all the attendant problems of interpretation. Nevertheless, a number of brave souls have attacked this problem. The details are beyond the scope of this review but we are interested in certain aspects of the experimental techniques employed.

The identifying feature of double beta decay is the emission of two beta particles in exact coincidence and (in the case of no neutrino emission) with a unique energy sum. Most attempts to observe double beta-decay have, therefore, been based on the observation of coincidences.

Fireman (56) reported an apparently positive result on Sn^{124} using coincidences between a pair of thin window Geiger-Müller counters. A half-life of about 10^{16} yr. seemed indicated. This result was not confirmed by Kalkstein & Libby (57) using a double screen-wall counter, who set a lower limit of 10^{17} yr. on the half-life. Fireman & Schwartzer (58) using cloud chamber photographs found a similar lower limit.

The studies of McCarthy on Sn^{124} and Zr^{96} (59) and on Ca^{48} (60) introduced a determination of the disintegration energy. The detectors used were a pair of organic scintillators (stilbene crystals in one experiment, plastic scintillators in the other) placed on opposite sides of the sample. Anticoincidence shielding by larger stilbene crystals or by a GM umbrella eliminated coincidences due to penetrating charged particles. The apparatus was placed underground beneath a large building for further shielding. The energies of coincident pulses were summed electronically and displayed on a charter recorder for later analysis (rates were so low that a multi-channel analyzer was not necessary, although a 30-channel analyzer was used in the first experiment). Background was thus reduced to about 0.2 c./hr. in a 1-Mev wide channel at 4 Mev. By these techniques a sensitivity was obtained capable of detecting a half-life of the order of 10^{17} yr. for double-beta decay of Ca^{48} when two samples of different isotopic composition were compared. An apparently positive result was reported.

Awschalom (61) failed to confirm these results and set lower limits of 2×10^{18} yr. on Ca^{48} and 0.5×10^{18} yr. on Zr^{96} using a similar apparatus. A lower background was obtained by working in a 1000-ft. deep salt mine.

The most recent attempt to observe double beta-decay is that of Cowan *et al.* (62) using liquid scintillation techniques. The sample investigated was enriched Nd^{150} for which the decay energy available is calculated to be about 4.4 ± 0.8 Mev. A 2 gm. sample of Nd_2O_3 was mounted between two liquid scintillation counters using 5 in. diameter photomultiplier tubes. The efficiency of the system for betas of the expected energy was about 30 per cent. This system was immersed in a 600 l. scintillation counter (using twelve 5 in. diameter photomultiplier tubes) run in anticoincidence with the sample counter to eliminate charged particle background. Shielding was 4 in. of Pb or Bi. The sum of the energies of coincident pulses was determined with a 100-channel pulse height analyzer. The ultimate sensitivity of this arrangement is such as to detect a half-life of the order of 10^{18} yr. No significant activity was observed although the calculated Majorana half-life (1.3×10^{15} to 6×10^{17} yr.) is considerably less than this limit.

One very great difficulty from which the double beta-decay methods have suffered is the apparent necessity of using isotopically enriched and depleted samples in pairs in order to make accurate correction for background. This greatly limits the sample size which can be used and hence plagues the studies with all the troubles of very long counts (days) at extremely low rates (a count per hr. or so). If an experiment could be devised in which the electronic analysis of the events was sufficiently detailed to identify double beta-decay and a large sample (several kg.) of the normal element used, relying on a neighboring element of similar Z and ρ for background, the sensitivity could be sufficiently expanded, and the half-life range might be extended to 10^{20} yr. or better.

An example of the sensitivity attainable is afforded by the work of Reines, Cowan & Goldhaber (63) who were able to established the minimum half-life of the proton as greater than 10^{21} yr. for possible nucleon-destroying collisions within the nucleus by analysis of the normal background of one of the neutrino detectors. It is of considerable interest that such an extreme limit could actually be established. The secret of such a great range is the extremely large number of protons contained in 300 l. of toluene, namely 2.3×10^{4} moles of H. The sample is, therefore, 10^{6} larger than in an experiment using say a few gm. of a medium atomic number nuclide. Granted that the proton is a particularly easy nuclide to incorporate into large scintillation solutions, it would nevertheless seem worth considering the possibility of incorporating really large samples of possible double beta emitters, such as Sn, Zr, etc., into large scintillators.

The Neutrino

Perhaps the most difficult of all low-level counting problems is the actual detection of the neutrino; the hypothetical particle whose existence is necessary to preserve the conservation laws during beta decay (64). Probably no one doubts the existence of the neutrino, which is urgently required by the fundamental laws of physics in their present form. However, the *ad hoc* character of all its properties are unsatisfactory and the actual observation

of an event caused by a neutrino at some point distant from the site of its formation would be very reassuring. In addition, as mentioned under double beta-decay, there remains the question of whether or not neutrinos and anti-neutrinos constitute a Dirac pair.

The one event which it is certain must be caused by neutrinos is the inverse of beta-decay. The normal decay of a neutron results in a proton, a negative electron, and a neutrino. If a neutrino of this type and of sufficient energy were captured by a proton, the conservation laws would permit this decay to go the other way resulting in the emission of a positron and the formation of a neutron. The cross-section for the neutrino capture process can be calculated. Unfortunately, it turns out to be alarmingly small: for fission neutrino capture by protons, the expected value is about 6×10^{-44} cm.^{2} or 6×10^{-20} barn. (It has been suggested that a new unit of cross section of about this size might be called the "shed.") The magnitude of this number can better be appreciated by noting that the entire earth is a very "thin foil" to neutrinos and that the neutrinos produced in the solar reactions escape from the enormous mass of the sun with practically no attenuation. Neutrinos have been called "the ashes of the universe" since once they are produced in nuclear reactions there is little hope of their energy ever being converted to any other form.

In spite of this unpromising situation, several methods of detecting neutrinos have been proposed. Only one has as yet come close to producing a positive result, namely, the project of Cowan & Reines. That project proposed (65) to observe directly the neutrino-proton interaction by placing near a large nuclear reactor a liquid scintillation counter of sufficient size to permit a detectable number of proton decays to occur. With the neutrino flux available just outside the shielding of a large Hanford reactor, Reines & Cowan calculated that a counting rate of several tenths of a c./min. might be obtained by using a liquid scintillation counter of 300 l. volume. Because of the extremely large background of such a detector, every possible means had to be used to select those counts due to neutrino capture. Fortunately, neutrino capture in a large scintillator initiates a complex and almost unique sequence of events. The capture results in the immediate emission of a positron with an energy of several Mev (assuming fission product neutrinos). The positron loses this energy and is annihilated almost instantaneously giving two 0.5-Mev gamma rays. The neutron is moderated to thermal energies and is captured by hydrogen in the normal scintillation solution. Since the capture time by hydrogen is long (hundred of μsec.), it is advantageous to add to the solution some Cd (in the form of Cd propionate, or better octoate). The capture time is then shortened to a few μsec., and a total of 9 Mev of gamma-rays in cascade (average multiplicity 3 to 5) is emitted (46). The plan in the experiment was, therefore, to look for an original pulse (positron plus annihilation gammas) with the proper energy (2 to 8 Mev.), followed within a few μsec. by the neutron capture pulse. The energy spectrum of the first pulse was determined with a 10-channel analyzer and the time delay of the second pulse

measured with an 18-channel time delay analyzer (0.5 μsec. channel widths). Elaborate shielding, including water, borax-loaded paraffin and Pb, was used around the detector, as well as an anticoincidence umbrella of GM tubes. Even with these precautions a pile-independent delayed coincidence background of about 2 c./min. was observed. This effect was ascribed to fast neutrons produced by μ meson capture in the shielding, by "stars," etc. The difference in this delayed coincidence signal with pile on and pile off (i.e., with and without a neutrino flux) was 0.41 ± 0.20 c./min. compared with a predicted rate of $\sim\frac{1}{3}$ c./min. on the basis of the 6×10^{-20} barn cross-section (66, 67). The most significant aspect of this result is that the counter sensitivity did indeed approach that required for a conclusive measurement. The experiment is currently being repeated at the DuPont Company at Savannah River, Georgia, under the auspices of the AEC, using an improved detector.[2]

One of the principal difficulties encountered in large detectors is the adequate collection of the scintillation light. Since it is difficult to cover more than a small fraction of the tank wall with phototubes (4% for the Los Alamos neutrino experiment counter; ninety 2-in. photomultipliers, 10 cu. ft. tank) the light must undergo a large number of reflections before reaching a phototube surface. Under these conditions it can be shown (68) that the fraction S of emitted light reaching a phototube surface is a function like $S = F/(1-rt)$, where F is the fraction of the wall surface covered by photomultipliers, r is the reflectance of the wall and t is the transmission of the solution for the average light path between reflections. It is clear that this function is extremely sensitive to the value of the product rt, especially when F is small. Therefore, the solvents used for large detectors must have an extremely small absorption coefficient over the scintillation spectrum (mean paths of 10 m. or more) and the wall reflectivity must be very high. Both of these ends are considerably easier to attain if the emission spectrum of the scintillator is shifted as far as possible to longer wavelengths. POPOP (peak emission at 4300 Å) is particularly useful as a secondary solute for this purpose, since terphenyl and 2,5-diphenyloxazole (the common primary solutes) emit at 3500 and 3800 Å, respectively.

The problem of wall reflectivity is made more difficult by the necessity of immersing the reflector in the solution. Since the reflectivity of a pigment depends on the refractive index difference, immersion in liquids of high refractive index destroys the reflectivity of otherwise adequate pigments of low refractive index, such as MgO. The only pigment combining high refractive index with reasonable reflectivity seems to be anatase TiO_2 of selected grades. Rutile TiO_2 is poor and some grades of anatase are inadequate. Even the best anatase fails rapidly for wavelengths shorter than 4000 Å. Specular reflectors (such as Al, Cr, Ag) do not approach the best pigments in total reflectivity. For long, narrow counters with phototubes at the ends, total internal reflec-

[2] The successful completion of this experiment was announced in June, 1956 [Reines *et al.* (70)].

tion at transparent walls is probably the best system but such an arrangement is difficult for large detectors with their attendant mechanical problems caused by large mass.

The problems of the selection of proper solvent, the incorporation of other elements (such as Cd in the neutrino experiment), lead too far afield to be discussed here. Large liquid scintillators are only beginning to be understood and their advantages exploited. There is considerable room for progress and many problems in low level counting and cosmic ray and fundamental particle research will benefit from their use.

An alternative and very ingenious method of neutrino detection was originally proposed by Pontecorvo in 1936, elaborated by Alvarez in 1949, and finally attempted by Davis in 1954 (69). In this method the neutrino is to be captured by Cl^{37} causing negatron emission leading to A^{37}. A^{37} is a particularly easy nuclide to detect in small quantities. It is noble gas and can thus be easily separated from extremely large quantities of carbon tetrachloride (the Cl-bearing chemical usually chosen). It decays by electronic capture with a 34 day half-life and is detected by the highly converted x-ray. Davis' experiments were carried out at the Brookhaven reactor at power levels of 20 to 25 Mw., using 36 to 75 day exposures. The neutrino flux through the CCl_4 was calculated to be 4.4×10^{11} sec.$^{-1}$ cm.$^{-2}$. CCl_4 volumes of 200 and 3900 l. were used. The A^{37} was measured in anticoincidence shielded Geiger-Müller counters. The results were negative and placed an upper limit of 2×10^{-42} cm.^{2} on the cross section. The limiting factor was the production of A^{37} by cosmic ray reactions in the CCl_4. This experiment is being repeated at the Savannah River Pile along with the Cowan & Reines experiment. The combination of the two results should be extremely interesting, since if neutrino and anti-neutrino differ, the CCl_4 experiment will give a negative result while the proton-neutron conversion will be positive. The CCl_4 method, on the other hand, can be expected to detect the solar neutrinos (since these are emitted during positron decay processes) if the C-N cycle is the source of solar energy. The *p-p* cycle neutrinos have an energy below the Cl-A threshold. However, the sensitivity of even a 3900-l. tank is a factor of 1000 below the calculated value necessary to detect the solar neutrino flux even for a pure carbon-nitrogen cycle source.

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NUCLEAR REACTORS FOR ELECTRIC POWER GENERATION[1,2]

LEON DAVIDSON, WILLIAM A. LOEB, AND GALE YOUNG
Nuclear Development Corporation of America, White Plains, New York

INTRODUCTION

Controlled nuclear chain reactions involving neutron-induced fissions can be established in a variety of combinations of materials (1). The simplest chain reacting structure is exemplified by the Los Alamos Scientific Laboratory's critical assemblies "Godiva" and "Jezebel" which are bare solid spheres of U^{235} and Pu ^{239} respectively, of the order of several inches in diameter (2).

Such solid spheres of fissionable material do not lend themselves to practical use as power sources because of heat removal considerations. To allow production of reasonable amounts of heat power without creating excessive internal material temperatures, it is necessary to pass a coolant through the chain reacting zone. This coolant might be a liquid metal with little slowing down effect on the neutron velocity, or it might be a hydrogenous liquid such as water or an organic compound. The coolant in this latter case slows down the neutrons, acting as a "moderator" and tending to thermalize the assembly. Since thermal reactors[3] have certain advantages over fast reactors, (for example the ability to operate with uranium fuel containing only small percentages of U^{235}), many concepts have been developed in which the chain reacting assembly includes, besides fuel and coolant, additional material whose primary function is to serve as a neutron moderator. High-purity graphite, beryllium, water, and heavy water are most often considered for this purpose.

The simple chain reacting sphere of pure fissionable material may thus be transformed conceptually into a fairly elaborate heterogeneous power producing structure, in which a coolant flows through channels in a structure of fissionable material and moderator. It is usually desirable to separate the coolant from the other materials by walls of some structural material. These sometimes take the form of pipes or ducts through which the coolant flows, and sometimes the form of sheathing or cladding on individual pieces of fissionable or moderator material. An individual piece of fissionable material,

[1] The survey of literature pertaining to this review was completed in March, 1956.

[2] Abbreviations used in this paper are: BTU = British Thermal Unit; gpm = gallons per minute; hp. = horsepower; kw. hr. = kilowatt-hour; mr = milliroentgen; psi = pounds per square inch; psia = pounds per square inch, absolute.

[3] A "fast" reactor is one in which essentially all of the fissions are caused by fast neutrons (in the Mev range). A "thermal" reactor is one in which the bulk of the fissions are caused by thermal neutrons (with energies about 1/40 ev). An "intermediate" reactor is one in which most fissions are caused by neutrons in an intermediate energy range (say from 1 to 1000 ev).

with or without some associated structural diluent or moderator material, clad or sheathed with an inert structural metal, is often called a "fuel element" and the typical reactor which has been conceptually described above is called a "solid fuel" reactor.

Another conceptual approach to reactor design is typified in its simplest form by still another Los Alamos Scientific Laboratory device, the "water boiler" (3). (This is a misnomer, for the device does not ordinarily operate in the boiling range.) The LASL water boiler is essentially a homogeneous chain reacting body consisting of a sphere of water about one foot in diameter in which sufficient U^{235} has been dissolved to achieve criticality. A "reflector" of graphite surrounds this to reduce the number of escaping neutrons. To allow continuous operation at a significant power level, heat must be removed from the fuel solution. In the prototype LASL device, this is accomplished by a cooling water coil located in the spherical container. However, the heat on occasion has been removed not by the cooling water but by allowing the fuel solution to boil.

A reactor of the type just described could be called a "liquid fuel" reactor. If the fuel is contained in a dilute aqueous solution the reactor will be thermal, with the fuel solution acting also as the moderator. Another concept of a "liquid fuel" reactor involves the use of a nonmoderating liquid (for instance a liquid metal, such as bismuth) to carry the fuel. This would not in general be a thermal reactor unless a separate moderator material were provided, for example graphite or beryllium.

The physical properties of the materials used as moderators, coolants, fuels, and structural materials will determine a great many of the characteristics of a reactor. Material characteristics are often found to be controlling factors when attempting to convert a conceptually promising materials combination into an engineered reactor design.

In short, power producing reactors may be considered to be composed of a wide variety of materials arranged in a wide variety of ways, although not all of these are sensible. As Weinberg (4) says, "The central issue in reactor development is to trace out of this welter of possibilities the dozen or so which are most likely to succeed." In the following pages we will attempt to show the situation as it stood at the beginning of 1956. Many different types of power reactors had been proposed for electric power generation, and a good many were actually under construction in several countries around the world (5). It is impossible to say whether one of these particular reactors represents the type which may some day dominate the power reactor field as the gasoline engine now dominates the automobile field. It is interesting to consider that eventually other types of nuclear power reactors might displace the first widespread types, much as the Diesel locomotive has recently captured the locomotive market in the United States. Since the ultimate selection cannot be known now, it is of interest to examine examples of the major types now being developed.

EXAMPLES OF POWER REACTORS

During 1955, the technological feasibility of generating electricity from nuclear reactor heat was demonstrated on many occasions and in many places.[4] In the United States, the public electric supply of the town of Arco, Idaho (pop. 1200) was derived entirely from the Borax-III reactor at the National Reactor Testing Station, for one hour late at night on July 17, 1955 (6). On July 18, 1955, at West Milton, N.Y., the Submarine Intermediate Reactor (SIR) MkA for a three hour period delivered about 4 megawatts of electric power into the local power company's lines (7). Near Moscow, Russia, a 5 megawatt atomic electric power station started operation in June, 1954 (8). Further examples of electricity production were furnished by the submarine propulsion reactors, the STR MkI reactor at the National Reactor Testing Station, Idaho and the STR MkII reactor installed in the U.S. Submarine "Nautilus," although the electricity produced by these reactors was not delivered to power lines for public use (9). The small French power reactor, G-1, started operating in 1956.

It is evident that these activitives did not in any way demonstrate feasible economic nuclear power. However, other nuclear reactor developments in 1955 gave more promise of leading eventually to economically competitive reactors. In the following pages we describe about twenty five examples of such reactors. These include designs which are actually under construction, as well as some which are in advanced stages of conceptual design and development work.

The recent growth in this field has been so rapid that it is not feasible to attempt to list all of the reactors now under serious consideration. The omission of several worthwhile reactor projects from the following discussion is due only to space limitations. It is nevertheless hoped that a sufficient number of examples of the major reactor types have been included to give a fair picture of the current situation.

Information is given on the basic features of the reactor examples as they were known early in 1956. In cases where several examples are very similar, not all details have been included for each. The current literature should be followed for up-to-date information, since many of the proposed designs reported here are subject to change as development work and construction proceed.

Table I below serves as an index to the reactors to be covered, and gives a brief description of each reactor. The reactors are discussed in more detail following Table I.

[4] The first demonstration of the generation of electricity from nuclear reactor heat took place at the National Reactor Testing Station in Idaho in 1951, when steam from the EBR-1 (a sodium-cooled experimental breeder reactor) was used to operate a 200 kw. turbo-generator. Another early example was the HRE-1 (an experimental aqueous fuel solution reactor) at the Oak Ridge National Laboratory, which generated 150 kw. of electricity in 1952. These demonstrations did not deliver power for external public use.

TABLE I

Examples of Power Reactor Designs Being Considered in 1955

Example Number	Reactor Name and Location	Owner	Operating Date	Electric Output, Mw.
	Solid Fuel, H_2O Moderator, H_2O Coolant (Non-Boiling)			
1	Army Package Power Reactor (APPR-1), Ft. Belvoir, Va.	U. S. Army	1958	1.3 (plus steam for heating) or 2.1 (no steam)
2	Brussels Exposition Reactor, Brussels, Belgium	Private Belgian Syndicate	1958	11.5
3	Consolidated Edison Reactor, Peekskill, New York	Consolidated Edison Co. of New York	1960	140 (plus 110 from oil superheater)
4	Pressurized Water Reactor (PWR), Shippingport, Pa.	U.S.A.E.C. and Duquesne Light Co.	1957	100
5	Yankee Pressurized Water Reactor, Rowe, Mass.	Yankee Atomic Electric Co.	1960	134
	Solid Fuel, H_2O Moderator, H_2O Coolant (Boiling)			
6	Experimental Boiling Water Reactor (EBWR), Lemont, Illinois	U.S.A.E.C.	1956	3.7
7	Closed Cycle Boiling Reactor, Elk River, Minn.	Elk River Rural Co-op. Power Assoc.	1959	22
8	Dual Cycle Boiling Reactor (NPG), near Chicago, Ill.	Nuclear Power Group (Commonwealth Edison)	1960	180
	Solid Fuel, D_2O Moderator, D_2O Coolant (Non-Boiling)			
9	Nuclear Power Demonstration Reactor (NPD), Des Joachims, Ontario	Atomic Energy of Canada, Ltd. and Power Comm. of Ontario	1960	20
	Solid Fuel, D_2O Moderator, D_2O Coolant (Boiling)			
10	D_2O Boiling Reactor, (U.N. Paper 495)	(Proposed by Argonne Nat'l. Lab.)	Not Scheduled	250
11	Halden Reactor, Halden, Norway	Institutt for Atomenergi	1958	10 to 20 Mw of heat (No electricity)
	Solid Fuel, D_2O Moderator, Gas Coolant			
12	Gas Cooled D_2O Moderated Reactor, U.S.S.R.	U.S.S.R.	Uncertain	100
	Solid Fuel, Graphite Moderator, Gas Coolant			
13	Calder Hall Reactors (eight), Calder Hall, Cumberland, and Annan, Dumfrieshire, Great Britain	United Kingdom Atomic Energy Authority	1956	About 46 per reactor
14	G-2 Reactors (two), Marcoule, France	French Atomic Energy Comm.	1958	About 30 per reactor
	Solid Fuel, Graphite Moderator, H_2O Coolant (Non-Boiling)			
15	Atomic Power Station (APS-1) near Moscow, Russia	U.S.S.R.	1954	5
	Solid Fuel, Graphite Moderator, Sodium Coolant			
16	Sodium Reactor Experiment (SRE), Santa Susana, California	U.S.A.E.C. and North Amer. Aviation	1957	7.5
17	Consumers Public Power Reactor, Columbus, Nebraska	Consumers Public Power District	1959	75
	Solid Fuel, No Moderator, Sodium Coolant			
18	Developmental Fast Breeder (APDA), near Detroit, Mich.	Power Reactor Development Co.	1960	100
19	Dounreay Fast Reactor, Dounreay, Scotland	United Kingdom Atomic Energy Authority	About 1960	About 15
20	Experimental Breeder Reactor (EBR-II), Arco, Idaho	U.S.A.E.C.	1959	15
	Liquid Metal Fuel, Graphite Moderator, Circulating Fuel Coolant			
21	Liquid Metal Fuel Reactor (LMFR)	(Proposed by Brookhaven Nat'l. Lab.)	Not Scheduled	210
	Aqueous (H_2O) Slurry Fuel, Self-Moderator, Cooling by Circulation of Slurry			
22	Suspension Power-Only Pile, Arnhem, The Netherlands	Reactor Centre Netherlands and N.V. "KEMA"	Uncertain	About 10 to 15
	Aqueous (D_2O) Slurry Fuel, Self-Moderator, Cooling by Boiling of Slurry			
23	Boiling Homogeneous Reactor, U.S.S.R.	U.S.S.R.	Uncertain	About 50
	Aqueous (H_2O) Fuel Solution, Self-Moderator, Cooling by Circulation of Solution			
24	Forced Circulation Power Reactor Experiment (LAPRE-I), Los Alamos, New Mexico	U.S.A.E.C.	1956	2 Mw of heat (No electricity)
25	Natural Convection Power Reactor Experiment (LAPRE-II), Los Alamos, New Mexico	U.S.A.E.C.	1956	1 Mw of heat (No electricity)
	Aqueous (D_2O) Fuel Solution, Self-Moderator, Cooling by Circulation of Solution			
26	Homogeneous Reactor Test (HRT), Oak Ridge, Tenn.	U.S.A.E.C.	1956	0.3
27	D_2O Fuel Solution Reactor	(Proposed by Oak Ridge Nat'l. Lab.)	Not Scheduled	100

DETAILS OF INDIVIDUAL REACTOR DESIGNS

The following paragraphs give information about the reactor examples listed in Table I. Some of the information presented here is based on private communications.

For each major type of reactor a general description will first be given, followed by individual reactor descriptions. (See Table III for the cost of construction.)

Solid Fuel, H_2O Moderator, H_2O Coolant (Non-Boiling)

This type of reactor (usually called the "pressurized water" type) makes up the largest segment of the U.S. power reactor program, and is also being used in the U.S. naval propulsion reactor program (10). Much development work has been done on this reactor system, and the first large scale power reactor scheduled to operate in the United States (the PWR at Shippingport, Pa.) is of this type.

"Pressurized water" reactor designs generally have the following features:

(*a*) The fuel is in solid metallic form having large surface per unit mass. Flat fuel plates about 0.060 inch thick may be used, arranged in elements consisting of about six to twelve parallel plates, spaced about 0.060 inch apart. These elements are typically about four inches square and about six feet long (exclusive of end fittings). The fuel is clad with a corrosion resistant material such as zirconium or stainless steel.

(*b*) The primary coolant is water (H_2O) flowing between the parallel fuel plates. The primary water circuit is pressurized to prevent boiling, and transfers its heat to a secondary water circuit, operating at a lower pressure, in which steam is formed.

(*c*) The primary water which flows between the fuel plates also serves as the neutron moderator.

To get reasonable thermal efficiencies, the secondary steam conditions are usually set somewhere above 480°F., 570 psia. The necessary temperature differential between the primary and secondary circuits in the heat exchanger (about 50°F.) requires the primary water circuit to be at least 530°F., and a minimum primary pressure of 890 psia is then required to prevent boiling. Actually in order to guard against boiling at localized hot spots and at localized low pressure regions (cavitation), an overpressure of 500 to 1000 psi is usually provided. Hence pressures of the order of 2000 psia are used in the primary circuits, making necessary thick-walled pressure vessels around the reactor cores.

The use of the primary coolant (H_2O) as the moderator requires some enrichment of the uranium fuel. Hence "pressurized water" reactors have been developed and used chiefly in the United States, where enriched fuel is readily available.

Example No. 1. The Army Package Power Reactor (APPR-1).—This is a military reactor to provide electric power and steam at remote installations, and is small in comparison with civilian power reactor designs (11, 12). Its

heat output is 10 Mw, and its gross electric power generation is about 2.1 Mw., of which about 10 per cent is used to operate the power station equipment. At this electrical output, there is little steam available for outside use.

The fuel elements in this reactor contain UO_2 particles dispersed in thin stainless steel plates, clad with stainless steel. The uranium initially is 17.7 per cent by weight U^{235}. A "burnable poison," boron carbide, is also included in the fuel plates. The B_4C, by absorbing neutrons, requires a high initial loading of U^{235}, but as the boron becomes consumed its poisoning effect lessens, roughly balancing the U^{235} burnup. By this means, a total burnup of 7.5 kg. of U^{235}, equivalent to 15 megawatt-years of operation (1.5 years at full power) is planned without refueling.

The primary coolant is water at 1200 psia, which leaves the fuel region at 450°F., and passes through a vertical U-tube steam generator which has a stainless steel clad steel shell and stainless steel tubes. The primary water then is pumped back through the reactor. Superheated steam formed in the secondary circuit (407°F. and 200 psia) is fed to the turbine.

The reactor is surrounded by an iron and water shield about four feet thick (made in sections suitable for air transport) outside of which is ordinary concrete. The radiation dose is reduced to one-fifth of the allowed tolerance level at the control room.

Example No. 2. The Brussels Exposition Reactor.—This was the first power reactor to be sold by private American industry to a foreign country (13). It is intended to produce 11.5 Mw. gross electrical output, and will be featured at the International Universal Exhibition in Brussels, Belgium in April, 1958. As it is a commercial venture, few details have been made available. It will be owned by syndicates of private Belgian concerns.

Zirconium fuel element cladding is to be used. Non-boiling H_2O will be the moderator and primary coolant, and a secondary H_2O circuit will produce steam in U-tube heat exchangers to drive the turbine. The fuel will be uranium containing a few per cent of U^{235}, to be supplied by the U.S.A.E.C. in accordance with an American-Belgian "bilateral agreement" on atomic energy (14).

Example No. 3. The Consolidated Edison Reactor.—Two features of this reactor proposal (15) are the use of thorium in the fuel region and the use of an oil-fired steam superheater.

The fuel consists of highly enriched uranium (90 per cent U^{235}) alloyed and clad with Zircaloy, forming plates 0.060 inch thick, about 2.5 inches wide, and about six feet high. These vertical plates are stacked alternately with 0.080 inch thick vertical Zircaloy-clad metallic thorium plates ("fertile plates") leaving 0.100 inch wide channels for upward coolant flow. About 210 kg. of U^{235} and 8100 kg. of thorium are contained in the fuel loading.

The fuel and fertile plates form an active "core" region six feet high and six feet in diameter, surrounded by a six inch thick water layer (neutron reflector) and steel thermal shields. All this is encased in a cylindrical steel

pressure vessel about 10 feet in diameter with walls 7 inches thick, designed for 1800 psi pressure with the walls at 550°F. The vessel is clad inside with stainless steel 0.1 inch thick. Outside there is glass thermal insulation, surrounded by a larger steel tank filled with water for gamma and neutron shielding. The water depth above the pressure vessel reduces the gamma dose to less than 7.5 mr/hr. during full power operation.

The maximum fuel surface temperature is about 600°F. Primary coolant water enters the core at 480°F., and leaves at 510°F. To prevent boiling, 1500 psia pressure is maintained in the primary circuit. The average heat flux at the fuel element surfaces is less than 200,000 BTU/hr. ft.^{2} The total flow of primary coolant is about 120,000 gpm. The heat exchangers are shielded because of the 6 Mev gammas coming from the 7-second N^{16} formed in the coolant water by the (n, p) reaction.

In the heat exchangers, preheated feed water at 332°F. and 420 psia is converted to saturated steam at 449°F. at a rate of about 1,900,000 lb/hr. at full power (500 Mw. of heat). The primary coolant flows through U-tubes, and the secondary steam forms outside the tubes. The steam is superheated to 1000°F. in an oil-fired superheater (200 Mw. heat rating) and then flows to the turbines. The gross electric output of the plant is 236 Mw. and the gross efficiency is about 34 per cent.

It is planned to replace only one-third of the core every four months. An initial excess reactivity of 8 per cent is allowed for overcoming burnup. The xenon transient is alloted another 8 per cent excess reactivity, the change from ambient to operating temperature 2.4 per cent, and the equilibrium Xe and Sm about 3.4 per cent. About 5 per cent extra negative reactivity for shutdown is provided. The control rods, which will be either boron-containing stainless steel or hafnium, therefore equal a total of 27 per cent of reactivity. There will be one central regulating rod and twenty shim rods, each separately driven through interlocked controls.

The discharged fuel elements will be stored for an indefinite period, and no reprocessing plans have been announced.

Example No. 4. The Pressurized Water Reactor (PWR).—The PWR design, described by Simpson & Shaw (16),was the first large plant to be authorized in the United States and will be owned by the U.S.A.E.C. The PWR will initially have a nominal heat output of 230 Mw., a gross electric power generation of 68 Mw., and a net station output of 60 Mw. The turbogenerator is being designed for 100 Mw. electrical output, however, since improvements in fuel element design will eventually permit increases in the operating power.

Mandil & Palladino (16) discuss the philosophy of the mechanical design of the fuel region of a PWR-type reactor. They indicate that the PWR itself will be a developmental reactor, with extensive instrumentation within the core to measure temperatures and flow rates during operation. They point out the need for experimentation during the design period, to solve the many

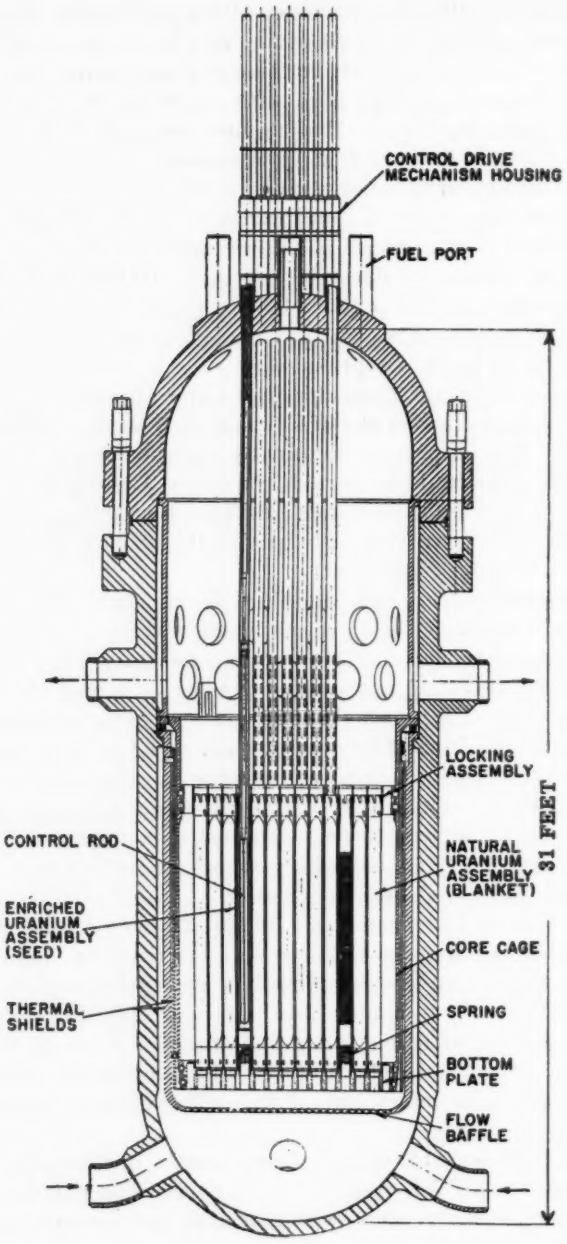

FIG. 1. Vertical section through pressure vessel of a pressurized water reactor.

engineering problems which cannot be answered analytically. Donworth & Lymon (16) examine in detail the operation of the PWR power plant in conjunction with a metropolitan electrical system.

The PWR fuel region will contain 15 tons of natural UO_2 as sintered cylindrical pellets, loaded into Zircaloy tubes about 0.4 inch in diameter. (The center temperature of the UO_2 in some tubes may reach 2200°F., while the surfaces of the tubes will not exceed 630°F.) The fuel region is a cylinder about six feet in diameter and six feet high.

To make the reactor critical a "seed" fuel region is provided, containing 75 kg. of highly enriched uranium alloyed with zirconium and formed into plates clad with Zircaloy-2 (0.070 inch overall plate thickness). The seed region forms a vertical cylindrical annulus about six feet high, three feet in diameter, and six inches thick, lying within the UO_2 fuel region. Natural UO_2 fuel is thus located inside and outside of the seed region.

The core is located in a pressure vessel, about 33 feet high and nine feet in inside diameter, with walls about nine inches thick (See Figure 1.) The top of this vessel is held on by 42 threaded studs each six inches in diameter to resist the 2000 psi internal pressure. Primary coolant water flows at 45,000 gpm upward between the fuel rods and fuel plates, serving both as moderator and coolant. It enters the core at 508°F. and leaves at 542°F., with surface heat fluxes of the various fuel elements being about 40,000 to 400,000 BTU/hr. ft.^{2} The heated primary water passes through stainless steel steam generators where it forms secondary circuit steam at about 600 psia and 486°F. for the turbine.

The insulated pressure vessel is surrounded by a shield consisting of three feet of water. A fuel burnup equivalent to about 0.3 per cent of the total uranium may be achieved in the UO_2 fuel, while the enriched uranium "seed" fuel is to last for 3000 hours of operation at full power. It is possible to rearrange the fuel elements periodically to equalize burnup or to increase the core reactivity attainable from the remaining fuel.

Example No. 5. Yankee Pressurized Water Reactor.—As revised late in 1955, the design for this reactor calls for about 780 kg. of U^{235} (at an isotopic concentration of 2.7 per cent in uranium). The heat output will be 480 Mw., giving 134 Mw. of gross electrical output. The reactor in general probably will resemble the PWR type as dicussed in Example 4.

Solid Fuel, H_2O Moderator, H_2O Coolant (Boiling)

One of the drawbacks of the reactors described above is the need for expensive high pressure construction to prevent boiling of the moderator-coolant. When the stability of boiling moderator reactors was indicated by the "Borax" experiments (17) it became feasible to proceed with the design of large reactors in which the fuel elements are cooled by the boiling moderator liquid. The steam from the fuel region may be used directly in a turbine. Therefore, a boiling reactor pressure vessel would only have to withstand the pressure utilized in the turbines. (See Example No. 6 below.)

The radioactive contamination of the turbines and other equipment needing maintenance should not be any worse in a boiling reactor plant than in a pressurized reactor plant if an intermediate steam circuit is used. If the steam goes directly to the turbines, there would be need for some provision to prevent carryover of radioactive solid particles or solutes arising from corrosion. The possibility of leakage of fission products from defective fuel elements also tends to favor use of a secondary steam circuit for driving the turbines. The heat transfer is good enough in these two-circuit boiling systems to allow reasonably small temperature and pressure differences between the two circuits, resulting in primary pressures much less than in the nonboiling water reactors. (See Example No. 7, below.)

The fuel regions of these reactors resemble those of the pressurized water types, but are somewhat larger, because the spacing of the fuel elements must provide for a larger upward volume flow of the primary coolant, which now consists of a steam and water mixture. The effect of bubble formation on the moderator properties depends on the particular design details. In most cases the reactivity decreases as the bubble volume increases. The effect which this has on the design of controls for these reactors will be discussed below. (See Example No. 2.)

Example No. 6. The Experimental Boiling Water Reactor (EBWR).—This reactor (scheduled to start up in 1956) is of interest as the first operating boiling reactor power plant designed for the purpose (18). (The Borax-3 power demonstration (19) was not really designed as a power plant.) The EBWR uses a direct cycle in which the H_2O steam formed in the fuel region flows directly to the turbine. No intermediate heat exchanger is needed and there is no secondary coolant loop.

The H_2O which cools the fuel elements also serves as moderator. Two interchangeable types of fuel element are used, one containing natural uranium, the other containing slightly enriched uranium (about 1.4 per cent U^{235} in the uranium), to enable adjustment of the U^{235} loading in the fuel region. The calculated total loading requirement is 66 kg. of U^{235}, including that in the natural uranium.

The fuel is uranium metal alloyed with a few per cent of other elements. Six Zircaloy-2 clad fuel plates form a fuel element, with channels 0.37 inch thick between plates for coolant flow. About 110 elements form the reactor core, which is four feet in diameter and four feet high. This is contained in a seven foot diameter pressure vessel, 23 feet high, made of stainless steel clad steel about 2.5 inches thick.

The coolant circulates by natural convection up through the fuel channels and down in the annular space between the core and the pressure vessel wall. The average steam content in the coolant channels is about 20 per cent. The steam from the channels rises through four feet of water and enters the "steam dome" formed by the upper portion of the pressure vessel. Here the entrained water droplets are removed by gravity. At the design power level of 20 Mw. (heat), 60,000 lb./hr. of saturated steam at 488°F. (615 psia) are

produced, most of which goes directly to the condensing turbine. The condensate at about 110°F. is returned to the annular water space surrounding the core.

There is no automatic adjustment of reactor power to match steam demand. Instead, power is set manually by the nine hafnium shim-safety rods. The steam pressure is automatically controlled, however, through a bypass valve which admits steam directly to the main condenser. As the turbine steam demand decreases the bypass valve opens. To allow for small rapid upward fluctuations in power demand without changing the shim rods, it is planned to allow a nominal 5 per cent of the steam to be continually by-passed. Increases over 5 per cent in power demand will require shim rod changes, while any decrease in demand will open the bypass further.

The reactor and steam system are enclosed in a sealed steel containment shell 80 feet in diameter and 119 feet high. The reactor is shielded by three inches of lead and about seven feet of concrete. Gross electric output is about 4.8 Mw., and net output is about 4.5 Mw. (electrical). The designers feel that the power can eventually be increased to 40 Mw. heat, and 9 Mw. electrical. They have planned the equipment so that D_2O may be used instead of H_2O as the coolant and moderator. A fuel element burnup of about 1.1 per cent of the total uranium atoms was used as a basis for cost estimates, and no decisions were reported on fuel reprocessing.

Example No. 7. The Closed Cycle Boiling Reactor (Elk River).—The designers of this reactor (20) propose it as suitable for plants of 10 to 20 Mw. electric output, but feel that for much larger plants it would not be competitive with the pressurized water type of reactor.

The reactor is similar to the EBWR design (Example No. 6). The primary steam in this case (about 1200 psia and 567°F.) does not go directly to the turbine. Instead, it condenses in the tubes of a boiler, producing saturated secondary steam at about 650 psia and 500°F. This may be superheated by an oil-fired unit to about 825°F. and 615 psia, which are typical conditions for the small modern steam power plants with which this plant design might compete.

This plant is intended to be simple to operate and easy to maintain. The use of a secondary steam loop keeps the radioactive primary steam out of the turbine, reducing maintenance problems. The intermediate boiler is elevated so that the primary condensate from the boiler tubes flows back by gravity to the reactor, eliminating the need for pumps or valves in this radioactive circuit. Control and stress problems are simplified by holding each loop at constant temperature and pressure. The primary system pressure is held by automatic movement of the control rods. The secondary system pressure is held constant through a feature of the intermediate boiler design. By varying the secondary water level in the horizontal drum, the length of the vertical primary steam tubes immersed in the water is varied. Since the secondary steam production rate is proportional to the submerged tube area, it can thus be controlled as needed.

The use of the secondary steam loop requires a temperature differential in the boiler which raises the needed pressure in the reactor vessel from about 600 psi, as in the EBWR (Example No. 6) to about 1200 psi, as used here. This is one penalty of this design, and tends to make the reactor construction more expensive than for the EBWR type. However, the absence of radioactivity from the secondary circuit should reduce plant maintenance costs.

Example No. 8. The Dual Cycle Boiling Reactor (*NPG*).—The most interesting feature of this design is the way in which the power is made to respond to the demand (21).

For stability, the core design of a boiling reactor usually provides that reactivity decreases as the percentage of steam bubbles increases. This feature is useful in an experimental installation since it makes the reactor inherently safe; an upsurge of power leads to increased boiling and decreased reactivity. However, this situation is not desirable in a power plant, since sudden increases in demand for steam will lower the pressure, increase the boiling rate, and decrease the power, whereas it should be increased.

The Nuclear Power Group reactor solves this control problem by the ingenious application of a dual steam cycle. The main steam supply comes directly from the core region, at 541°F. and about 970 psia, and is fed to the high pressure entry of a dual pressure turbine. A secondary steam supply at 467°F. and 500 psia is fed to another inlet of the turbine. Turbine load variations operate a governor valve which controls the secondary steam. The secondary steam may come from a "flash tank" operating at 500 psia pressure, into which 970 psia water from the reactor is admitted through the automatic valve. About 12 per cent of this water flashes into secondary steam, while the residual water, cooled to 467°F., is pumped back into the reactor, where it cools the boiling water, reduces the total bubble volume, and thereby increases the reactivity. When the automatic regulating valve increases the production of flash steam, it also increases the rate of "cool" water return to the reactor, leading to the desired increased reactor power. Instead of a flash tank, the 500 psia steam may come from a secondary circuit in a boiler heated by 970 psia water, admitted through a regulating valve from the reactor. The 970 psia subcooled water returning to the reactor will reduce the reactor temperature and increase the reactor power.

Many details of the core design for this project are not readily available. Slightly enriched uranium will be used, in the form of UO_2 clad with Zircaloy-2. A burnup of 1.1 per cent of the uranium is the design goal. The reactor will produce about 700 Mw. of nuclear heat, yielding 192 Mw. gross electric output and 180 Mw. net electrical output. Pumping power requirements will be about 9 Mw.

Solid Fuel, D_2O Moderator, D_2O Coolant (Nonboiling)

Example No. 9. Nuclear Power Demonstration Reactor (*NPD*).—The only announced example of this type of reactor is being built in Canada (22). The

moderator D_2O is held at a temperature in the range 60°F. to 260°F., and is thermally insulated from the fuel and coolant. This permits the moderator temperature to be adjusted to provide reactivity changes as part of the control scheme.

The fuel loading consists of roughly five tons of natural UO_2 or uranium metal as round rods clad with Zircaloy. These are suspended in vertical channels in a stainless steel D_2O tank about 12 feet in diameter. The system pressure is 1200 psia. A D_2O reflector region one foot thick surrounds the ten foot diameter fuel region. Primary coolant D_2O flows through the fuel channels, reaching 530°F. without boiling, and then going through shell-and-tube heat exchangers. Here, saturated H_2O steam is formed at about 450°F. and 420 psia to drive the turbine. The nuclear heat production rate is about 86 Mw., the gross electric power is about 20 Mw., and the net electrical output is 18 Mw.

No mechanical control elements are required, all the usual functions being handled by the D_2O moderator. For operating control the moderator level in the tank will be varied. For shim control (long-term changes to compensate for reactivity lost by fuel burnup, etc.) the temperature of the moderator will be changed. The safety control is provided by dumping D_2O moderator from the core region.

Solid Fuel, D_2O Moderator, D_2O Coolant (Boiling)

A boiling D_2O reactor allows more freedom in core design and fuel enrichment than is possible with an H_2O boiling reactor. For example, the D_2O cooling channels in the fuel region may be thicker than would be used for H_2O coolant.

The D_2O steam from the fuel region could be condensed in a boiler which generates H_2O steam for the turbines. (This scheme would be similar to Example No. 7.) The D_2O steam may instead flow directly to the turbines, as in the proposed D_2O boiling reactor (Example No. 10). It may be difficult, however, to keep D_2O uncontaminated as it passes through the turbine and condenser. The design of a leakproof vacuum condenser, large in size and with low temperature drop, may present a serious problem.

Example No. 10. D_2O Boiling Reactor.—Iskenderian *et al.* (23) propose a boiling D_2O reactor of 1000 Mw. heat output, yielding 270 Mw. gross electric output and 248 Mw. net electric output, using natural uranium fuel. It is indicated that this type of reactor would be uneconomic at ratings much less than 1000 Mw. heat output, although the use of slightly enriched uranium would permit consideration of reactors having only about 250 Mw. heat output (60 Mw. electric output).

The fuel would be about 44 tons of natural uranium metal in the form of parallel plates 0.11 inch thick, clad with Zircaloy-2, with 0.350 inch channels between plates. A fuel element contains nine such plates, and about 300 such elements form a vertical fuel region, 12 feet in diameter, located in a pressure vessel 16 feet in diameter and 45 feet high. About 100 tons of liquid D_2O in

the lower half of the pressure vessel comes to three feet above the fuel elements, and also provides a D_2O side reflector around the fuel region. The upper half of the vessel serves as a steam dome.

D_2O at about 485° F. and 600 psia flows up through the fuel elements, becoming about 80 per cent steam by volume. The steam flows out through the steam dome, and the remaining liquid flows down an annulus outside the reflector layer, and out to the pumps. The D_2O condensate returning from the turbine condenser at 90°F. is fed to the spaces outside the 300 fuel element tubes to lower the temperature of the moderator, improving nuclear performance and preventing moderator boiling.

The fuel may reach 1 per cent burnup of total uranium if limited by net reactivity loss only. No estimate of the metallurgical limit of the life of these fuel elements is given. There was no indication by early 1956 that any direct-cycle boiling D_2O solid fuel reactor would be built in any country.

Example No. 11. Halden Reactor.—Dahl (24) has reviewed the Norwegian and Dutch situation with regard to power and ship propulsion reactors. A design study was made for a non-boiling pressurized D_2O natural uranium reactor similar to the Canadian Nuclear Power Demonstration Reactor. There are severe problems in maintaining a 300°F. temperature difference between the moderator D_2O and the coolant D_2O. The probability of availability of slightly enriched fuel led the Norwegian-Dutch group to consider a small boiling D_2O reactor, producing steam at about 445°F. With such high D_2O moderator temperature, a natural uranium power reactor of this size might not become critical, so that the reactor will be "spiked" with a few enriched elements. The fuel would be hollow uranium cylinders, about 4 inches long, 1 inch in outside diameter and 0.4 inch in inside diameter, contained in vertical aluminum-nickel alloy tubes. About nine tons of uranium and fifteen tons of D_2O will be needed. The reactor vessel would be a vertical cylinder about 9 feet in diameter with an effective fuel region height of about 8 feet.

About 40,000 lbs. of H_2O steam per hour (300 psia pressure) will be made in heat exchangers in which D_2O steam condenses. The D_2O condensate will return by gravity to the reactor tank, eliminating need for a pump. The H_2O steam will be used in a nearby paper mill as process steam, and will not be recirculated. Reactivity control is provided by two operating rods and eleven shim-safety rods, containing a cadmium alloy. No electrical generation is planned at this plant, but it is a possible prototype of an industrial or central heating reactor, and the design could be used for power generation.

Solid Fuel, D_2O Moderator, Gas Coolant

Example No. 12. Gas Cooled D_2O Moderated Reactor.—Vladimirsky (25) points out that in D_2O moderated reactors, the fuel may have a high surface-to-volume ratio, because large amounts of cladding can be tolerated. This permits high power output per unit mass of fuel. There is, however, the

penalty of the high cost of D_2O. The designer must minimize both the holdup and the losses of heavy water to get a useful design.

A D_2O moderated D_2O cooled nonboiling reactor was first considered, with a secondary circuit for H_2O steam. However, the holdup of D_2O in the heat exchanger circuits would lead to a total D_2O inventory of about one ton per Mw. of electrical power, which was considered excessive. The use of light water coolant would not have reduced this figure, since the power would have decreased.

A gas-cooled reactor design was therefore developed in which the cylindrical D_2O moderator tank is pierced by vertical aluminum alloy tubes (fuel channels) through which CO_2 gas flows at about 600 psia pressure. The tube walls are cooled by the D_2O which surrounds them. In these tubes are suspended the natural uranium metallic fuel elements, in the form of thin rods or plates. The fuel cladding (unspecified) can withstand about 1000°F. maximum surface temperature. The CO_2 leaves the fuel channels at about 930°F., and passes through boilers which produce superheated steam (H_2O) at about 450 psia and 750°F., for the turbine.

The turbine is also fed some low pressure saturated steam (about 275°F., 45 psia), produced in the low temperature end of the CO_2 heat exchanger, to improve overall efficiency. About 100 Mw. (electrical) will be produced, of which about 15 per cent is consumed by the CO_2 circulation blowers. The heavy water inventory is about 0.35 tons per Mw. of electrical power.

Solid Fuel, Graphite Moderator, Gas Coolant

The gas-cooled graphite-moderated reactors which form the basis of the British and French power reactor programs are not much different in principle from the Oak Ridge X-10 air-cooled graphite reactor which started operating in 1943. They are designed to operate with natural uranium fuel which makes expensive isotope separation plants unnecessary. The use of gas cooling, rather than water, avoids the potential danger of a power surge which might occur if the cooling water (H_2O) were lost from a graphite-moderated reactor. The British emphasized safety in choosing this reactor type.

Gas-cooled reactors consume from 15 to 20 per cent of their gross electrical output in driving the coolant blowers. (In fact, the much smaller French air-cooled plutonium production reactor, G-1 (26) consumes about 8 Mw. of electrical power to drive its gas blowers, while producing a total of only about 6 Mw. of electricity from its generators.)

The CO_2 cools the clad uranium metal fuel cartridges which rest in channels in the graphite moderator. The hot CO_2 then passes through heat exchangers where superheated steam is generated.

Example 13. Calder Hall Reactors.—These reactors (27) are built in pairs, feeding their steam to a common generator house which contains two 23 Mw. turbogenerators for each reactor. Altogether, the four reactors which

are to be located at each reactor station of this type will have a combined gross electrical output of 184 Mw., but the net station output is reported to be only 125 Mw., so that almost one-third of the electric output is consumed within the reactor station. The variable speed CO_2 blowers alone consume about 6 Mw. per reactor, or roughly 15 per cent of the gross electrical output.

The Calder Hall reactors, consisting of about 1000 tons of graphite, are contained in steel pressure vessels about 60 feet high and 40 feet in diameter, with walls two inches thick. The CO_2 gas, at about 115 psia pressure, flows upward through channels in the graphite moderator.

The reactor contains about 40 tons of natural uranium fuel rods, clad in finned tubes of an unspecified metal. These heat the CO_2 to about 600°–700°F. before it flows to the heat exchangers. There are four of these per reactor, each a vertical steel cylinder about 70 feet high and 18 feet in diameter, containing 2 inch diameter steel boiler tubes (with studded surface on the gas side to improve heat transfer), divided into two circuits. A high pressure circuit heated by the incoming CO_2 produces superheated steam at about 200 psia for the main supply to the turbines, while a low pressure circuit makes use of the heat of the exiting CO_2 and delivers low temperature steam at 50 psia to the turbine secondary inlet.

Since one of the primary functions of these reactors is to produce plutonium for Great Britain's atomic weapon and fast reactor programs, a condenser is provded which can handle all of the steam, permitting full power operation even if the turbines are inoperative. The fuel charging provisions are located on the top of the reactor, along with the drives for the boron steel control rods. Future improvement may be possible through the use of slightly enriched uranium fuel, which would permit higher temperature cladding materials to be used, and thus enable higher thermal efficiencies to be attained.

Example No. 14. G-2 Reactors (France.)—These reactors like those at Calder Hall, have the dual functions of producing plutonium and electric power (28). They, therefore, do not run at the high temperatures needed to achieve high efficiencies of power production. The heat output of each G-2 reactor is 150 Mw., the gross electrical output about 37 Mw., and the net electric output 30 Mw.

The pressure vessel, which is over 30 feet in diameter and over 30 feet long for each reactor, is made of thick prestressed concrete which serves also as the shielding. A cooled steel liner keeps the concrete cool. The graphite moderator has 1200 horizontal fuel channels, about 3 inches in diameter, in which 100 tons of uranium is loaded. The fuel slugs, about one inch in diameter and one foot long, are natural uranium alloyed with a little aluminum. They are contained in finned magnesium cans which are limited to 750°F. external temperature, while the central uranium temperature is held to about 1000°F. maximum. (The second limit governs in the high-flux central region of the reactor, while the first limit governs in the low-flux outer portions of

the fuel region.) The CO_2 coolant gas, which flows through the fuel channels at a pressure of about 220 psia, reaches about 570°F.

Fuel may be charged and discharged from one channel at a time while the reactor continues full power operation. A gas-tight charging device inserts a fresh fuel rod into the CO_2 outlet side of the fuel channel. The fuel rod at the other end falls down a chute, then through a retarding device to the reactor exit lock. Elimination of the need for refueling shutdowns will improve the power and plutonium output of these reactors.

Solid Fuel, Graphite Moderator, H_2O Coolant (Nonboiling)

This type of reactor is similar to the Hanford plutonium production reactors of the U.S.A., which are graphite moderated and cooled by water flowing through "process tubes" which contain the fuel elements. However, to prevent boiling in the coolant channels, at the higher temperatures used, the coolant pressure must be on the order of 2000 psia. (Boiling which occurs in a channel which has been designed for non-boiling operation may lead to reactor instability or to "boiling burnout," which is a form of damage involving melting of the fuel elements and injury to the process tube.) The fuel channels must therefore be made of a high strength material rather than aluminum as used at Hanford. This usually results in a need for partial enrichment of the uranium fuel.

These reactors do not, however, require the large thick walled pressure vessels needed by the pressurized water reactors. (Example Nos. 1 to 5.) The high coolant pressure is contained by the small diameter process tubes, and the massive graphite moderator need not be enclosed in a heavy pressure vessel.

Only one reactor of this type is discussed in detail below, because of space limitations. Another interesting proposal of this type may be found in the paper by Andersen, Carson, & Love (29).

Example No. 15. Atomic Power Station (APS-1).—This small Russian nuclear power plant (8) was the first non-propulsion reactor to generate electricity in Megawatt quantities, having started up in June, 1954. It has a 30 Mw. heat rating, and generates 5 Mw. of electricity, using enriched uranium fuel with graphite moderator and pressurized water (H_2O) coolant. It is a developmental reactor, having instrumentation in the fuel region to study temperatures and flow rates, as well as having a thermal column and other research facilities.

The uranium used contains 5 per cent U^{235}, and is alloyed with an unspecified metal. It is formed into long hollow rods, jacketed externally with a flexible metal tube (unbonded) and internally with a stainless steel tube through which the pressurized primary coolant flows. The 128 vertical fuel channels in the graphite moderator structure form a cylindrical core region about five feet in diameter and six feet high, surrounded by a two-foot graphite reflector, all of which is enclosed in a sealed cylindrical steel jacket filled with helium or nitrogen.

The cooling water, at about 370°F. and 1500 psia, flows down a separate internal tube to the bottom of each fuel element, then flows up around this tube inside the fuel element itself. The heat transfer rate is about 500,000 BTU/hr. ft.^{2} The water leaves the top of the element at about 510°F. and flows to the steam generators where, in the secondary circuit, superheated steam is produced at about 180 psia and 490°F., for the turbine.

The outlet water temperature from the fuel elements is made uniform within about 15°F. by setting the proper water flow for each channel to compensate for the radial power variation. The fuel loading is changed every two months, the job taking several days. Fuel elements from the outside of the core are moved to the center, to increase burnup.

The cold reactor would go critical with 60 fuel elements. Eighteen boron carbide shim rods are used to permit loading all 128 fuel channels, the total fuel loading containg 27.5 kg. U^{235}. This provides excess reactivity sufficient to overcome xenon, temperature effects, and burnup. A burnup of 20 per cent of the U^{235} is achieved (through shuffling of the fuel) which results in a U^{235} concentration of 4 per cent at discharge. The initial conversion ratio (Pu^{230} atoms produced per U^{235} atom burned) is only about 0.3.

Water leakage from the fuel channels into the graphite is guarded against by automatic individual shut-off valves and other safety provisions. Two automatic regulating rods ordinarily hold the power level within 3 per cent of the desired value, and the steam conditions are quite constant. In the first year of operation, there were no fuel element failures and very little down-time for repairs.

Solid Fuel, Graphite Moderator, Sodium Coolant

The use of liquid sodium or a similar liquid metal (e.g., sodium-potassium alloy, "NaK") permits attainment of quite high temperatures at low working pressures; for example, 1500°F. can be attained in liquid sodium without exceeding atmospheric pressure. Since liquid sodium technology is now well advanced (30), this coolant can be considered for application in power reactors. Satisfactory materials of construction, pumps, handling equipment, and the like, are becoming available commercially.

It is considered advisable to prevent the sodium from coming into contact with the graphite, so that the moderator blocks are canned in metal in present designs. Fuel elements may be clad with stainless steel.

One disadvantage of sodium is the radioactivity induced in it by neutron activation (the 15 hr. Na^{24} gamma activity). The desire to avoid a possible explosive chemical reaction between H_2O and radioactive primary sodium leads to the use of secondary sodium circuits, receiving heat from the primary coolant in liquid metal-liquid metal heat exchangers. The secondary coolant then goes to a boiler in which steam is produced for the turbines.

Example No. 16. Sodium Reactor Experiment (SRE).—A discussion of this graphite moderated, sodium cooled reactor is given by Parkins (31). The SRE is actually an experimental facility, but, as now being constructed,

will generate 7.5 Mw. of electricity from the 20 Mw. (nominal) of nuclear heat.

The fuel will be uranium metal enriched to 2.8 per cent U^{235}, in the form of slugs 0.75 inch in diameter and 6 inches long, contained in stainless steel tubes having 0.100 inch wall thickness, with a 0.010 inch bonding layer of NaK between the slugs and the tube walls. A cluster of seven tubes forms a "fuel element"; about 37 such fuel elements will be needed to achieve the 20 Mw. (heat) power level. The total U^{235} loading is about 71 kg. Since fuel element development is one of the purposes of this program, no estimate of fuel life or burnup is available.

Each fuel element is located in a channel about 3 inches in diameter, in the center of a hexagonal graphite column about 11 inches thick. The graphite is canned on all surfaces with 0.035 inch thick zirconium. For reasons of gas pressure control, a 0.25 inch diameter vent tube connects the interior of each moderator column can to the helium space at the top of the reactor.

The graphite columns form a cylinder about 10 feet high and 10 feet in diameter which includes a 2 foot thick outer reflector region. The fuel region is 6 feet high and 6 feet in diameter. The moderator is contained in a stainless steel tank, 11 feet in diameter and 19 feet high. The whole reactor is located underground, which reduces the concrete required.

The primary sodium coolant enters the bottom of the fuel channels at about 500°F., and flows up to the sodium pool which fills most of the reactor tank above the moderator region. A blanket of helium gas covers the sodium surface. Sodium at about 960°F. flows from the pool to a U-tube heat exchanger where sodium in a secondary loop is heated to 900°F. The secondary sodium then flows to a steam generator which produces superheated steam at 600 psi and 825°F. for the turbine.

The sodium is circulated by variable speed centrifugal pumps with frozen sodium shaft seals. (These pumps are very much cheaper than electromagnetic pumps.) There are four control elements and four safety elements in which boron-nickel alloy is the poison material. The average reactivity control is roughly 2.5 per cent per element.

Example No. 17. Consumers Public Power District.—A 250 Mw. heat output, 75 Mw. electrical output, sodium cooled, graphite moderated, uranium fueled, nuclear power plant is described by Starr (32). This reactor is very similar to the Sodium Reactor Experiment, SRE (Example No. 16). (Although an alternate design using thorium-uranium alloy fuel is also given in the paper, it will not be discussed here.) The fuel consists of uranium metal, enriched to 1.8 per cent U^{235}. The complete fuel loading consists of 215 fuel elements, containing a total of 443 kg. U^{235}, and forming a vertical cylindrical fuel region 10 feet high and about 13 feet in diameter. A 2 foot thick layer of graphite, not containing any fuel, surrounds the entire fuel region as a reflector. The graphite assembly is contained in a stainless steel tank, about 18 feet in diameter and 25 feet high.

The primary coolant (sodium) enters the bottoms of the fuel channels at 500°F., and leaves at about 925°F. Sodium in a secondary circuit is heated to about 900°F., and flows to steam generators which produce superheated steam at 800 psi, 825°F.

Eighteen boron steel control and safety rods operate in stainless steel thimbles which seal them from the primary coolant. Surrounding the reactor tank is a six inch thick layer of steel slabs, which acts as a "thermal shield" to stop the bulk of the gamma rays which would otherwise overheat the concrete biological shield. There is thermal insulation between the thermal shield and the concrete. (The whole reactor is located below ground, with the surrounding earth reducing the required concrete shielding.) The thermal shield is heated by the gamma rays, etc., to temperatures ranging from 500°F. to 1200°F., while the concrete is kept below 150°F. by an organic liquid coolant circulating in tubes.

The initial excess reactivity is 2 per cent, and the initial conversion ratio is 0.72. A fuel burnup of about 0.3 per cent of total uranium is expected. The free surfaces in the primary sodium circuit are blanketed with helium at a nominal positive pressure to prevent air from reaching the sodium, since oxygen contamination makes sodium quite corrosive to the stainless steel equipment.

Solid Fuel, No Moderator, Sodium Coolant

Only a fast neutron reactor seems likely to attain in practice a positive breeding gain using the U^{238}—Pu^{239} breeding cycle (plutonium being fissioned to produce power and extra neutrons being caught in U^{238} to produce more plutonium). Liquid metal coolants (being non-moderating) are generally used. The low vapor pressures of liquid metals permit very high temperatures to be attained in the power cycle without high pressure problems. The advantages and disadvantages of liquid metal coolants were discussed above in connection with the graphite-moderated sodium-cooled reactors.

The Experimental Breeder Reactor, EBR-1, the first NaK-cooled fast breeder reactor of the U.S.A.E.C., went into operation at Arco, Idaho in 1951. It has demonstrated that power can be generated while breeding is taking place.

The major advantage of this type of reactor is the possibility of production of more fissionable material than is burned. The decrease in the amount of fuel to be purchased, and the income from sale of excess fuel which is produced, give this type of reactor its chief economic promise. This promise tends to be offset by the following problems:

(*a*) A high fuel investment is required (a large critical mass at a high enrichment) because fission cross sections are generally low at high energies.

(*b*) The fissionable fuel is relatively undiluted with structural or fertile material, and is therefore subject to very intensive radiation which limits its life in the reactor and requires frequent reprocessing.

Thus the fuel inventory charge and fuel reprocessing cycle costs will have a major effect on the economics of this type of reactor.

In November, 1955, during reactor kinetics tests on the EBR-1, an experimental power surge which was purposely started was allowed to get out of control through human error. Then, because some safety interlocks had been inactivated, the reactor core was severely overheated and perhaps partially melted. No personnel were injured, and there was no external damage.

Example No. 18. Developmental Fast Breeder (APDA).—As described by Amorosi *et al.* (33), this sodium cooled fast reactor is intended to be a developmental and demonstration reactor. It will consume highly enriched uranium fuel and will produce plutonium for sale. The design calls for more Pu^{239} atoms to be formed than the number of U^{235} atoms consumed, but since the chemical reprocessing details have not been worked out, it is uncertain how much net Pu^{239} will be available for sale.

The plant will produce 300 Mw. of nuclear heat, making 100 Mw. of gross electric power, and delivering 90 Mw. of net electrical output. The fuel region will be loaded with 485 kg. of U^{235}, in uranium containing 27 per cent U^{235} by weight. The uranium will be alloyed with small amounts of another metal and formed into thin rods or plates clad with zirconium. The rods will form a vertical fuel region about three feet long and three feet high.

Around the fuel region will be a "blanket" region about 2 feet thick, consisting of 44 tons of "depleted" uranium (containing less than 0.7 per cent U^{235}) canned in stainless steel tubes. In this blanket neutrons from the reactor core form plutonium, which is to be recovered by chemical processing when enough has accumulated. A steel tank 14 feet high and 10 feet in diameter surrounds the blanket. This tank has no top, but opens directly into an upper tank about 12 feet tall and 16 feet in diameter, containing 15,000 gallons of liquid sodium at about 800°F. Sodium at 550°F. is pumped into the bottom of the reactor tank, flows upward along the fuel and blanket rods (there is no moderator) and enters the pool at 800°F. From there, it flows by gravity to the intermediate heat exchanger, where NaK (sodium-potassium alloy) in a secondary circuit is heated to 750°F. The NaK then goes to a boiler where it produces superheated steam at about 615 psia and 730°F., for the turbine.

An airtight steel cylindrical structure, about 84 feet in diameter and 150 feet tall, sunk about 50 feet into the earth, encloses the reactor and entire primary circuit. This structure will prevent escape of products of a reactor accident. There are eight safety rods, containing boron carbide highly enriched in the B^{10} isotope, and one similar regulating rod. A fuel burnup of from 4 to 12 per cent of U^{235} atoms per cycle (1 to 3 per cent of total uranium atoms) is the design goal.

Example No. 19. Dounreay Fast Reactor.—The paper by Kendall & Fry (34) indicates that this "reactor" is really an experimental facility in which different designs of cores and fuel elements may be tried out. Liquid sodium,

circulated by 24 electromagnetic pumps in parallel loops, flows through a "pot" about 10 feet in diameter and 12 feet high, in which may be placed test cores which are about 2 feet high and 2 feet in diameter. The hot sodium then flows through several concentric-tube helical heat exchangers, where a secondary coolant (sodium or NaK) is heated. The secondary coolant then flows to boilers in which steam is formed for power generation.

Stainless steel is used for construction of the sodium circuits, and no valves are used. The shield around the reactor pot consists of a four foot layer of graphite, poisoned with boron compounds to absorb the slowed neutrons. A steel sphere about 135 feet in diameter with walls 1 inch thick surrounds the reactor and primary sodium circuit to contain the products of any accident.

Example No. 20. Experimental Breeder Reactor (EBR-II).—Barnes *et al.* (35), describe the EBR-II which will produce 60 Mw. (heat) and about 15 Mw. (electrical). The purpose of the EBR-II is to demonstrate the feasibility of one type of fast reactor system, not to produce electricity competitively. The undeveloped state of the plutonium fuel art has led to the use of highly enriched uranium fuel, containing 60 per cent by weight of U^{235}, instead of a plutonium fuel. (200 kg. of U^{235} is used.) The uranium is in the form of thin metallic rods or "pins" clad in stainless steel tubes roughly 0.25 inch in diameter and 2 feet long. These are grouped to form a vertical hollow cylindrical fuel region, roughly two feet in outside diameter, nine inches in inside diameter, and two feet long. This is surrounded on all sides by a blanket region roughly three feet thick, and the "hole" inside the cylindrical fuel region is filled with blanket material also. The blanket consists of depleted uranium rods clad in metal tubes, and the core and blanket together form a cylinder roughly seven feet high and seven feet in diameter, contained in a close fitting steel tank. This is surrounded by a shield layer consisting of a two foot thickness of graphite and boron contained in steel cans, to minimize neutron activation of nearby equipment.

The whole reactor and shield assembly just described rests inside a large, double-walled, steel "primary tank" roughly 25 feet in diameter and 25 feet high. In this tank are also located the main heat exchanger and fuel storage containers, and in the top of this tank is the electromagnetic pump which circulates the coolant. The primary coolant is sodium which fills the primary tank and thus covers the reactor to a depth of about ten feet. This permits fuel assemblies to remain submerged in sodium when being unloaded from the reactor, so that they do not overheat because of the high afterheat generation rate.

The electromagnetic pump, which is over twelve feet tall including the driving motor and generator, uses a current of about 250,000 amperes, and draws about 500 kw. The main sodium piping is not welded tight, but slip joints are used at the pumps and other equipment, and leakage of sodium to the large pool in the primary tank is tolerated. The pump takes its suction directly from the primary tank at 770°F. The primary sodium leaves the

reactor at 900°F., then flows into the heat exchanger and discharges back into the primary tank. Pressures in the primary sodium circuit are on the order of 50 to 100 psia. Secondary circuit sodium leaves the heat exchanger at about 880°F. and goes to a boiler where superheated steam is generated at about 850°F., and 1250 psia for the turbine.

An important feature of this proposal is the plan for a self-contained pyrometallurgical fuel reprocessing plant which would remove the bulk of the fission products and allow preparation of new fuel elements by remote methods. A fuel burnup of about 1 per cent of the total atoms in the fuel elements is expected. Operating control is provided by axial movement of certain fuel elements out of or into their regular positions.

LIQUID METAL FUEL, GRAPHITE MODERATOR, CIRCULATING FUEL COOLANT

Example No. 21. The Liquid Metal Fuel Reactor (LMFR).—The use of a circulating fuel solution instead of solid fuel elements would permit fuel reprocessing and removal of fission products to be put on a continuous basis, and would eliminate the problem of radiation damage which limits the life of solid fuel elements (46). Aqueous fuel solutions require high pressure construction for high temperature operation, but a liquid metal fuel solution would eliminate this pressure problem.

The selection of bismuth as the fuel solvent for the LMFR was a compromise. It does not have as high a solubility for uranium as would be desirable, and under neutron irradiation it gives rise to a troublesome radioactivity (138 day Po^{210}). Its high density also presents pumping and flow problems. Nevertheless, these problems have been attacked fairly successfully. A graphite structure appears to be feasible, and a thermal reactor design has been developed. The reactor was originally planned to be a breeder, consuming the U^{233} fuel which it produced, in order to reduce the net cost of operation. However, the problem of developing a suitable breeding blanket has led to the postponement of breeding plans. A small experimental U^{235}-fueled reactor (LMFRE) will probably be built in the near future to demonstrate the core technology.

Miles & Williams (36) give the design of a nuclear power plant of 500 Mw. heat output, 210 Mw. electrical output. Schomer *et al.* (37) go into the mechanical construction problems. The fuel is a solution of less than 0.1 per cent of U^{233} (or highly enriched uranium) in bismuth.

The reactor core is a vertical cylinder of graphite, five feet in diameter and five feet high. Fuel solution flows up through holes in this graphite at a rate of about 1500 tons per minute, entering at 750°F. and leaving at about 1000°F. It is degassed and then flows through a heat exchanger which heats sodium in a secondary circuit to 960°F. This sodium passes through a steam generator which delivers superheated steam (1250 psia, 900°F.) to the turbine. (See Fig. 2.)

The graphite reactor core is encased in a graphite vessel about six feet in diameter and 12 feet high. A breeder blanket region three feet thick sur-

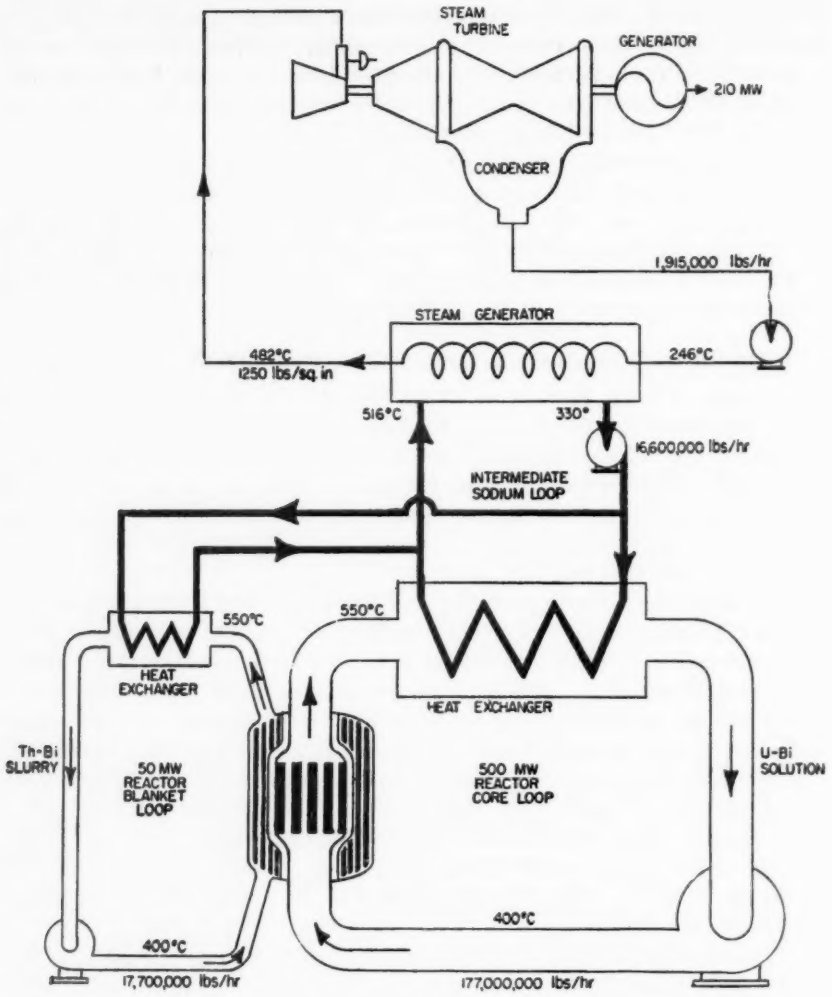

FIG. 2. Typical use of intermediate coolant loop between primary coolant and steam cycle. (This is a liquid metal fuel reactor.)

rounds this vessel. The blanket might be a slurry of thorium in molten bismuth, flowing upwards, moderated by graphite rods. About 50 Mw. of heat from the blanket is transferred to the sodium loop in a separate heat exchanger.

Both the fuel solution and the blanket slurry will be continually processed. Volatile fission products, as well as the polonium, come off as a gas. A sidestream of the fuel is extracted with fused salts to eliminate most fission products. Make-up uranium is added in this step. Remaining fission

products will be removed by batch reprocessing. Treatment with pure hot bismuth is used to remove the bred U^{233} from a sidestream of blanket slurry.

Aqueous (H_2O) Slurry Fuel, Self-Moderator, Cooling by Circulation of Slurry

Example No. 22. Suspension Power-Only Pile.—Aqueous slurry fuel reactors avoid many of the problems of solid fuels. The solid particles tend to settle out of a slurry unless there is continual turbulence, which may be provided by pumping the slurry through mixing chambers.

The data given below refer to a 250 kw. (heat) prototype design described by de Bruyn *et al.* (38). (A full scale reactor is planned to produce 50 Mw. of heat.) The fuel is a water slurry (H_2O) containing 4 per cent by volume of sintered UO_2 powder with particle sizes between 5 and 12 microns. This size permits the fission fragments to enter the liquid, while the Pu^{239} produced will remain in the particles.

About 2 kg. of U^{235} (in uranium enriched to 20 per cent U^{235}) will be needed for the prototype. (In the large reactor it is planned that D_2O will be used instead of H_2O, and the uranium will then need only slight enrichment.) The reactor core is a cylinder about one cubic foot in volume, in which the pressure is maintained at about 700 psia to prevent boiling at the 480°F. slurry exit temperature. Outside the cylinder is a reflector layer of BeO about six inches thick, followed by a graphite reflector several feet thick. The whole is encased in a steel vessel which maintains a hydrogen (or deuterium, if D_2O is used) pressure throughout the system, to reduce the required wall thickness of the core cylinder.

A gas separator removes Xe from the hot slurry and recombines the decomposed water. The slurry then goes through a heat exchanger. Operating control involves changing the UO_2 content and temperature of the slurry. The use of combination gas-lift pumps and heat exchangers is being considered for the full scale reactor. In the large reactor, constant processing of the water phase will remove a large fraction of the soluble fission products.

Aqueous (D_2O) Slurry Fuel, Self-Moderator, Cooling by Boiling of Slurry

Example No. 23. Boiling Homogeneous Reactor.—The use of a slurry of fuel particles in water (either H_2O or D_2O) may be combined with the "boiling reactor" concept. Advantages of slurry fuel and boiling coolants have been discussed in preceding examples.

The paper by Alichanow *et al.* (39) covers several related reactor types using D_2O or H_2O solutions and slurries of natural or enriched uranium fuels. An example which uses a suspension of finely divided UO_2 in boiling D_2O will be discussed. A thorium slurry in D_2O is used as a breeding blanket in this case. The fuel isotope may be U^{233} or U^{235}. The slurries are kept in suspension by the turbulence of the boiling action. After shutdown, after heat

generation or steam from an external source will provide sufficient boiling to maintain the suspension.

The boiling fuel slurry may fill only the lower part of the core pressure vessel, without organized circulation, while steam fills the upper half. The bubbles in the liquid decrease the fuel density and increase the critical mass needed. The reactor vessel may instead be filled with boiling slurry, which flows up to a steam separating vessel, from which the unvaporized slurry returns to the reactor core. In the latter case, the fuel holdup in the external circuit may be significant.

A steam purifier removes entrained water, uranium oxide particles, etc. A decontamination factor of about 10^{10} is reported, in tests of this device. A catalytic recombiner removes gases from water dissociation, and the steam then condenses in a heat exchanger. Condensate is added to the fuel slurry as it returns by natural convection from the steam separating vessel to the reactor. In the heat exchanger, secondary (H_2O) steam is produced for the turbogenerators.

Control characteristics have been calculated for this reactor with about 0.5 per cent UO_2 (by weight) in the slurry, operated at about 50 to 200 Mw (heat) with about 30 to 50 kg. of U^{233} held in the entire circuit. For constant slurry composition, the reactor power first increases with the pressure up to about 1000 psia (the bubbles become smaller) and then the power decreases to about zero at pressures about 2500 psia, because of the low liquid densities at the high saturation temperatures. A change in steam demand will change reactor steam pressure and produce the required change in power output. Operating control, to set the desired pressure-power relationship, may be achieved by throttling the slurry circulation.

Aqueous (H_2O) Fuel Solution, Self-Moderator, Cooling by Circulation of Solution

The two examples of this type of reactor come from the Los Alamos Scientific Laboratory, and use basically the same concept (40). A stainless steel cylinder about 15 inches in diameter and five feet high contains an aqueous phosphoric acid solution of highly enriched uranium. There is no external fuel circuit. The criticality is adjusted by sealed control rods or outside reflector movement. The fuel solution circulates past heat exchanger coils near the top of the vessel. (A cylindrical baffle separates the upflow and downflow.) Water is converted to steam in the coils, and this is condensed in an external heat exchanger which makes secondary steam. Radiolytic gas formation is held at an equilibrium value by the reverse reaction, at the temperatures and pressures used.

One of the major considerations in these reactors is the highly corrosive nature of the fuel solutions. Gold cladding (0.010 inch thick), a combination of silver and gold coatings, and platinum are used to protect the reactor parts. The corrosion problem is probably the main obstacle in the way of aqueous fuel solution reactors.

The design of both reactors reinforces the usual high negative temperature coefficient of reactivity of liquid fuel solution reactors, and provides stable operation. In these reactors, the space into which heated expanded liquid is forced effectively separates this portion of fuel from the chain reacting system. This serves to set an automatic upper limit to the average fuel temperature.

Example No. 24. Forced Circulation Power Reactor Experiment (LAPRE-I).—The fuel solution in this reactor (40) is made from phosphoric acid (7.5 Molar, containing about 50 per cent H_2O by weight). In this is dissolved 140 grams of uranium per liter. The uranium is enriched to about 90 per cent by weight U^{235}. At the 800°F. operating temperature, the vapor pressure is about 3900 psia, so that the 15 inch diameter reactor vessel has walls 3 inches thick. The wall thermal stresses induced by gamma ray heating limit the total power to about 2 Mw. (heat).

The fuel is circulated by a pump at the bottom of the reactor driven by a sealed rotor motor. About 8 kg. of U^{235} are needed in the fuel solution. Boron control rods in platinum-clad thimbles are used to shut down the reactor, while the upper power limit is set by the negative temperature coefficient. Increases in power demand on the intermediate boiling water circuit cause decreases in reactor fuel temperature which lead to increased reactivity and power output.

Example No. 25. Natural Convection Power Reactor Experiment (LAPRE-II).—This modification of the previous example is described by King (41). The essential change is the use of a fuel solution having a much lower vapor pressure. Highly concentrated phosphoric acid is used (17 molar, containing only about 10 per cent of H_2O). The vapor pressure is below 500 psia at 800°F. The uranium is kept in solution as the U^{++++} ion by means of a 200 psi hydrogen overpressure, which also prevents boiling of the fuel.

The operating pressure is much lower and the pressure vessel walls much thinner than in the LAPRE-I. This reduces the wall thermal stress and will eventually permit obtaining high powers in this type of reactor. The power of LAPRE-II is limited to 1 Mw. by the size of the internal heat exchanger used, and by the use of natural circulation of fuel.

The fuel of the LAPRE-II is less corrosive than the dilute fuel of the LAPRE-I, although gold cladding is still used on the internal stainless steel surfaces. A movable external graphite reflector is provided for shim control. The use of a burnable poison (boron) in the fuel solution would make the reactor completely automatic with no moving parts. A fuel life of several years and a power level of several Mw. might make this unit attractive as an unattended reactor.

Aqueous (D_2O) Fuel Solution, Self-Moderator, Cooling by Circulation of Solution

The two examples of this reactor type discussed here were developed by the Oak Ridge National Laboratory. (42, 43) Fluid fuel reactors have po-

tential advantage in fuel reprocessing and have definite control advantages. However, aqueous fuel solutions have proved to be highly corrosive (44). For example, the North Carolina State College reactor, after only two years of operation with UO_2SO_4 fuel solution in H_2O, had to be shut down for repairs (45).

The low cost fuel reprocessing methods to which these designs will lend themselves have not yet been demonstrated. However, the control advantages resulting from the high negative temperature coefficient of reactivity were established in tests on the earlier HRE-I reactor (42).

Example No. 26. Homogeneous Reactor Test (HRT).—As described by Beall & Swartout (42), the HRT is a developmental reactor with a heat rating of 5 Mw. It will use the turbogenerator equipment from HRE-1, and will produce about 0.3 Mw. of electricity.

The fuel solution contains about 4 kg. of uranium (containing about 90 per cent U^{235}) in the form of UO_2SO_4 dissolved in D_2O. The solution would contain about 1 per cent uranium by weight. It is pumped up through an approximately spherical Zircaloy-2 core vessel about 32 inches in diameter, in which its temperature rises from about 490°F. to about 570°F. An operating pressure of 2000 psia prevents boiling of the fuel solution. The fuel passes through a gas separator and heat exchanger, where H_2O steam (470°F. and 520 psia) is formed in a separate circuit. Some steam drives the turbogenerator, excess steam going to an aircooled condenser.

A liquid blanket region about one foot thick surrounds the core vessel. Liquid D_2O at 2000 psia will be used in this blanket as a neutron reflector, but eventually the blanket will contain a slurry of ThO_2 in D_2O so that some U^{233} can be bred. The blanket circuit contains pumps, heat exchanger, gas separator, etc., and when the thorium slurry is used, will contribute about 5 per cent of the total power.

The circuits described above are located in a shielded pit, which may be flooded with water to permit maintenance operations after a reactor shut down. There are no mechanical control or safety rods, dependence being placed on the high negative temperature coefficient of reactivity. The average temperature of the fuel during operation is set by the uranium concentration in the solution, which may be varied by evaporating D_2O from the solution or adding D_2O to it.

A system is provided for removing the non-condensable fission product gases and absorbing them on activated charcoal. No integral fuel reprocessing equipment is to be installed, but there will be a small pilot plant to develop continuous chemical extraction processes.

Example No. 27. D_2O Fuel Solution Reactor.—Briggs & Swartout (43) propose a large scale nuclear power plant with a gross electric output of 105 Mw. and a net output of 100 Mw., based on a reactor similar to the Homogeneous Reactor Test. The heat power would be 360 Mw. in the reactor core and 80 Mw. in the thorium oxide slurry blanket. The fuel would be UO_2SO_4 dissolved in D_2O. About 30 kg. of fissionable fuel isotopes (U^{235} plus U^{233})

and about 11 tons of D_2O are needed in the fuel solution. The blanket slurry requires about 16 tons of thorium and about 15 tons of D_2O. These fluids are passed through separate heat exchangers which produce saturated H_2O steam at 435°F. and 365 psia in a secondary circuit to drive the turbogenerator.

No control rods would be used, the aqueous fuel reactors being quite stable as discussed above. The production of U^{233} in the blanket slurry is expected to exceed the consumption requirement in the fuel region by about 60 grams per day, so that if low cost fuel reprocessing equipment could be incorporated into the plant, a very low net fuel cost might be achievable.

TECHNICAL AND ECONOMIC OUTLOOK

Trends in Power Reactor Technology

The power reactor designs described above illustrate the wide variety of approaches currently being followed. From this compilation, general technical trends may be discerned. Some of the most important of these are discussed below.

Fuel.—Most work to date has been on solid reactor fuels. In spite of this, no standard fuel element has yet emerged. It may be expected that such elements will soon receive increased emphasis. There is also a continuing development effort on fluid fuels.

Whether the fuel isotope will be U^{235}, Pu^{239}, or U^{233}, technologically is not too significant (48). Similar methods will eventually be used to handle any of them. To date, however, almost all efforts have been centered on U^{235}, at enrichments varying from natural concentration (0.7 per cent) up to about 90 per cent.

The manufacture of fissionable isotopes from U^{238} or Th^{232} has often been emphasized from a long-term resources viewpoint. As actual industrial interest in power reactors increases, it is also becoming clear that the conversion of fertile into fissionable isotopes (even if consumed within the same reactor, and not sold) is of major economic interest (49, 50). Thus, most reactors examined contemplate obtaining reasonably high conversion ratios using slightly enriched fuel. (In some cases, "seeding" or "spiking" of natural uranium fuels with a small proportion of highly enriched uranium in separate elements is found to be preferable to using a complete loading of partially enriched fuel.) Breeder reactors, producing more fissionable material than they burn, are of interest for the long run (51) but are, on the whole, not emphasized in today's program.

Moderator.—One of the major characteristics of a thermal reactor is the choice of moderator. H_2O, D_2O, and very pure graphite are the three moderators which have received major attention. There is also some effort aimed toward developing low vapor pressure, organic liquid moderators.

H_2O moderator has had the greatest attention in the U.S. and U.S.S.R. where enriched uranium is available. In other countries attention has cen-

tered largely on graphite and D_2O moderated reactors with natural uranium fuel.

Coolant.—Liquid coolants are widely used at present, although gas at moderate temperatures is used in several large reactors and some work is being done on high temperature gas coolants. H_2O is the most commonly employed coolant. There is, however, increasing activity in liquid metal and D_2O coolants. Countries other than the U.S. seem at present to favor gas or D_2O coolants.

Structural Materials.—The common structural material for use with water or liquid metal at high temperature is stainless steel. However, zirconium is now being used in many pressurized water reactors. Graphite is a popular structural material which, in the Liquid Metal Fuel Reactor (LMFR) design, will be used in contact with the molten fuel alloy.

The especially corrosive nature of many aqueous fuel solutions has caused an extensive search for materials to withstand attack by these solutions at moderate temperatures (47). No completely satisfactory material has yet been found for this application.

Equipment Components.—Many reactor components are now becoming available through commercial channels, both in the United States and in other countries. Pumps of the electromagnetic and canned rotor types, heat exchangers or boilers in which water may safely be heated by liquid sodium, and solid fuel elements of various types, all may be ordered from an increasing number of suppliers. Control rod drives, reactor instrumentation, and steel containment vessels are also items of commerce.

Economics

For more than ten years there has been an extensive effort to design, develop, and build nuclear power plants. Nevertheless, we are only now beginning to see actual cost data for power from such plants.

The cost of nuclear electric power may be divided roughly into three components. These are: Fixed cost, Operating cost, Fuel cost.

Fixed Cost.—This cost is ordinarily proportional to the capital investment in plant and equipment and is markedly affected by the length of the allowable amortization time. To estimate fixed cost it is important, then, to know the required plant capital investment and the plant life (52).

Required capital investment is particularly difficult to estimate for nuclear plants because of the large development expenses that have been required. Table II gives development costs for a number of representative units. Table III gives estimated construction costs excluding development. From this tabulation it appears that the cost of large units may reasonably be expected to be below about $250 per kilowatt of electrical capacity. This is a notable decrease from earlier estimates. Since this table is based on actual designs, it may be considered fairly reliable. It would be prudent, however, to wait until each plant is actually completed before considering any plant cost figures as definite.

TABLE II

APPROXIMATE DEVELOPMENT COSTS OF SOME POWER REACTOR DESIGNS
(In Addition to the Investment Costs Given in Table III)*

Example No.	Name of Reactor	Development Cost Millions of Dollars
4	PWR	$47
6	EBWR	16
16	SRE	9
18	APDA	15
20	EBR-II	24†
24	LAPRE-I	1.5
25	LAPRE-II	0.2‡
26	HRT	48§

* The HRT plant investment cost is $1,800,000. The LAPRE-I facility costs $700,000. The LAPRE-II costs $100,000. These are not given in Table III.

† This is exclusive of $6 million spent previously on the EBR-I program.

‡ In addition to LAPRE-I development costs.

§ This amount includes the full cost of the HRE-I (dismantled) and the program for developing large reactors (as in Example 27).

The plant amortization period is less well established. Until more experience has been amassed, it will not be possible to establish realistic plant lives. However, data are now becoming available for some nuclear reactors. The Hanford reactors have been in operation for twelve years, the ORNL X-10 research reactor for thirteen years, the EBR-I for five years, and the BNL research reactor for six years. None of these units are prototypes of the plants now being built, but they do give initial indications of a reactor life expectancy. We can, then, have confidence that nuclear plant lives of over ten years will be achieved. Whether power reactor plants will have thirty year lives has yet to be established.

Operating Costs (excluding fuel).—The operating costs include such items as labor, maintenance, health physics, and security. These items will undoubtedly be different from equivalent items for conventional thermal power plants. However, with the exception of maintenance, these are relatively predictable items and should not cause major uncertainties in the estimated electric generating costs. Maintenance is an item that could have major importance.

There is little doubt that reactor repairs can be effected. For instance, the Canadian NRX reactor and the North Carolina Research Reactor have had major repairs performed after long periods of operation. The high cost of working in the presence of radioactivity may, however, make this a major operating cost if extensive repairs are required. In the long run, when plant reliability is well established, it seems unlikely that this will be a major item of cost for nuclear plants.

TABLE III

ESTIMATES OF NUCLEAR POWER PLANT INSTALLED COSTS (INCLUDING TURBO-GENERATOR EQUIPMENT, BUT EXCLUDING DEVELOPMENT COSTS AND FUEL COSTS)

Plant	Example No.	Expected Operation	Total Cost, Millions of Dollars	Electric Power Capacity, Megawatts	Installed Cost of Plant, $ per Kw. (electric)
		Small Experimental Units			
EBR-2 (U. S.)	20	1959	15.3*	17.5	875
NPD (Canada)	9	1958	15	20.	750
EBWR (U. S.)	6	1956	3.6*	5.	720
SRE (U. S.)	16	1957	5.3*	7.5	710
Brussels Exposition Reactor	2	1958	5	11.5	435
		"Plutonium-Plus-Power" Plants (Government Owned)			
G2 (France)	14	1958	43	30	1430
Calder Hall (U. K.) (per single reactor)	13	1956	28	46	610
		Large Demonstration Reactors (U. S.)			
Fast Breeder (Detroit Edison Group)	18	1960	45	100	450
PWR (Shippingport, Pa.)	4	1957	37.7	100	377
Consumers' Public Power Dist. (Nebraska)	17	1959	24.3	75	325
Nuclear Power Group (Commonwealth Edison, Chicago)	8	1960	45	180	250
Yankee Atomic Electric Co.	5	1960	33.4	134	250
Consolidated Edison (Peekskill, N. Y.)	3	1960	55	250†	220‡

* Cost includes fabrication cost of first fuel loading.

† Includes 110 Mw. from use of oil-burning superheater.

‡ This low cost is not strictly comparable to the other cases listed, as almost half of the power in this case comes from conventional oil burning equipment, which is much cheaper than nuclear power equipment.

Fuel Cost.—The economic justification for nuclear power rests largely on the hope for fuel costs lower than those presently attained in fossil fueled plants. At present this is perhaps the largest area of uncertainty in the cost

of nuclear power. A large amount of work and a considerable amount of operating experience will be required before this cost category is well defined.

In order to achieve low fuel costs it will be necessary to burn an appreciable fraction of the nuclear fuel before it must be removed from the reactor system to be reprocessed or discarded. At the present time the major effort has been aimed at the development of solid fuel elements which can go to relatively high burnups. It is generally expected today that solid fuel elements can be made which will withstand burnups of approximately 1 per cent of the total atoms present in the alloy. The resulting fuel costs are still quite high and serious efforts will be required to raise this allowable burnup. Whether such efforts will succeed is still subject to considerable uncertainty.

An additional fuel cost is associated with the processing of spent fuel. The costs for this reprocessing are very much in doubt at present and represent a very large fuel cost uncertainty. This uncertainty will apply to the fluid fuel reactors as well as those with solid fuels.

Summary.—In summary, then, it is still too early to make accurate predictions of the power generation costs from nuclear power plants. There are in the literature many estimates ranging from about 6 mills per kwh to 20 mills per kwh. Careful refinement of these seems to be beyond our real capabilities at the moment. We have tried to outline above the major uncertainties that presently govern these estimates. It is clear that we are coming much closer to realistic cost estimates than we were able to do in the past. However, some plants will have to be operated and a number of major uncertainties will have to be resolved before these cost estimates can be considered reliable. If these uncertainties are resolved in a favorable fashion, it would appear that nuclear power will compete favorably in many locations with conventional thermal generating plants (53, 54).

CONCLUSION

To summarize the present-day state of the art, there are under construction or serious planning a large number of nuclear power plants. These range from small experimental units to full scale power plants. They cover many different types of technology and are to be designed and constructed by many different groups. Over the next five years nearly all of these plants should be in operation. From them will come large amounts of information and experience which will indicate the extent to which nuclear power plants can be expected to compete with other sources of electrical power. Already improved data on capital investment requirements for such plants are becoming available. Additional information of this sort will continue to appear over the course of the next few years. It can be expected that a version of this summary paper written in 1961 will contain more realistic estimates of the cost of nuclear power as well as actual data on nuclear reactors then in use for electric power generation.

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CELLULAR RADIOBIOLOGY[1,2]

By L. H. Gray

British Empire Cancer Campaign Research Unit in Radiobiology, Mount Vernon Hospital, Northwood, Middlesex, England

GENERAL INTRODUCTION

I. RADIOBIOLOGY OF THE CELL

1. The Size and Structure of Elementary Particles
   (a) *Transforming principle;* (b) *Viruses*
2. Investigations Employing Physical Variables
   (a) *Effects of individual particles;* (b) *Action spectra;* (c) *Microwave analysis and chemical change;* (d) *High intensity and ultra fractionation;* (e) *Drying, freezing, and low temperature*
3. Cellular Reactions to Nuclear and Cytoplasmic Injuries
4. Investigations Concerned with Cell Metabolism
   (a) *General metabolism;* (b) *Nucleic acid metabolism*
5. Some Questions Relating to the Production of Chromosome Aberrations
6. The Influence of Environmental Factors on the Response of Cells to Radiation
   (a) *Water content;* (b) *Oxygen;* (c) *Chemical pre-treatment;* Investigations in which treatment is reported to decrease the effectiveness of an irradiation; Investigations in which an enhancement of radiobiological damage has been reported; (d) *Investigations concerned with post-irradiation treatment;* Chemical and metabolic factors; Photoreactivation
7. The Influence of Linear Energy Transfer (LET) on the Biological Effectiveness of Ionizing Radiation
8. The Influence of Fractionation
9. Radiation Carcinogenesis
10. The Mutational Process and Genetic Damage

II. RADIOBIOLOGY OF CELLS AND TISSUES

1. Bacteria and Thymocytes
2. The Cellular Elements of Testis and Ovary
3. The Developing Embryo
4. Skin and Vascular System
5. Thyroid and Orbital Glands
6. Nerve, Retina and Muscle
7. Animal Tumours
8. Germination
9. Unclassified

[1] The following abbreviations have been used in this chapter: T.P. (transforming principle); DNA (deoxyribonucleic acid); RNA (ribonucleic acid); kmc (kilomegacycle); ATP (adenosine triphosphate); RBE (relative biological effectiveness); kvp (kilovolts, peak); kr (kiloroentgen).

[2] The writer is indebted to Dr. O. C. A. Scott for assistance in screening the literature to be reviewed.

This literature survey follows that of Mortimer & Beam (238) and attempts to cover papers published in the open literature during the calendar year 1955. Special reports from individual laboratories on work carried out under A.E.C. contract are not referred to. Abstracts of these reports generally appear in *Nuclear Science Abstracts*. A considerable part of British work in the field of radiobiology is reported currently in the *Annual Report of the British Empire Cancer Campaign*. In view of the exceptionally international character of the Geneva Conference on Peaceful Uses of Atomic Energy held during the year under review, advantage was taken of the fact that Volume 11 (*Biological Effects of Radiation*) of the Proceedings of this Conference was at hand at the time of writing, to refer to papers which were presented on Cellular Radiobiology. Titles and authors of some papers on Cellular Radiobiology presented at the meetings of the Division of Biological Sciences, Association of the Academy of Sciences of the U.S.S.R. on the Peaceful Uses of Atomic Energy, July 1 to 5, 1955, and since published (in Russian) are given in an appendix to the bibliography (370 to 380).

During the year there has appeared a second edition of Lea's book *Actions of Radiations of Living Cells*. It differs from the first edition only by minor editorial changes and additions from Lea's own notes. In his *Douglas Lea Memorial Lecture*, Coulson (67), who was one of Lea's early collaborators, reviews some aspects of Lea's work, particularly that concerned with bacterial inactivation. There has also appeared Volume II of Hollaender's *Radiation Biology* (152). This volume, which like the first is excellently conceived, presents authoritative accounts of a variety of topics outside the scope suggested by its sub-title: *Ultra-Violet and Related Radiations*, for example chapters which cover work with ionizing as well as ultraviolet and other radiations on *The Effects of Radiation on Protozoa and the Eggs of Invertebrates Other than Insects* by Kimball, *Radiation and Viruses* by Luria, *Effects of Radiation on Bacteria* by Zelle & Hollaender, and *Radiation Studies on Fungi* by Pomper & Atwood.

Cellular Radiobiology has been considered to include a study of the effects of irradiating organised tissues as well as cells, but not the whole organism, at least when this is a mammal. This makes a convenient if not entirely logical division; the question has already arisen as to whether a paper on the radiation syndrome in the Drosophila fly qualifies for inclusion. A much more serious problem arises as to how the selected material should be presented. The developmental pattern of radiobiology is of the greatest interest when seen as a whole, but confronts the reviewer, who must perforce trace it thread by thread, with a very difficult problem in classification. The following compromise has been adopted. In Section I, of this chapter, papers are grouped according to the problems which they attempt to illuminate and are mainly concerned with some aspects of the mode of action of radiation. In Section II, the remaining papers are grouped in relation to classes of cells or tissues whose reaction to irradiation is described. At the end of each group

in this section cross reference is made without further comment to all papers in Section I which have reference to this particular class of cells.

I. RADIOBIOLOGY OF THE CELL

1. The Size and Structure of Viruses and Macromolecules

Radiobiology has not infrequently added significantly to our knowledge of fundamental biological processes. This is true of a number of papers included in this review and is the aspect of radiobiology emphasized in experiments by Pollard (270 & 271) and his co-workers.

(a) *Transforming principle.*—

Pollard' and Ephrussi-Taylor & Latarjet (94), have used ionizing radiation to probe the nature, properties, and size of the active unit of the pneumococcal transforming principle (T.P.). Marmur & Fluke (226) have bombarded dry samples of pneumococcal DNA transforming principle with 2 Mev electrons. They find that when the irradiated material is assayed with respect to three drug resistant factors and one metabolic factor, the results show no difference in radiation sensitivity of the DNA associated with these four factors. Ephrussi-Taylor and Latarjet find T.P. to be extremely susceptible to attack by radicals formed by the radiolysis of water, and only in the presence of about 10 per cent yeast extract is an inactivation curve obtained which is believed to represent effects resulting from the direct ionization of the T.P. molecule. On the basis of the inactivation doses observed under these conditions the effective molecular weight is estimated not to exceed 700,000. This would be the weight of the material which must be ionized to cause loss of biological function of a single particle of T.P. The presence of oxygen at the time of irradiation does not enhance indirect inactivation. Drew (84) finds that Type 3 pneumococcal T.P. loses its biological activity when exposed in 0.85 per cent saline at pH 7.1 to 10,000r and upwards of ^{60}Co gamma rays. In the presence of 0.5 per cent cysteine at pH 7.1, however, loss of biological activity was only observed after 100,000r. Moreover, this dose of radiation delivered to the bacteria from which the T.P. was derived induced no measurable loss of biological activity in the T.P. subsequently extracted from them.

(b) *Viruses.*—

One investigation in this group concerns the relationship between colicine and bacteriophage. Colicine K and Phage T-6 are closely related as regards their host ranges. Latarjet & Fredericq (206) have measured the direct inactivation doses for each, and find that they are comparable. This fact suggests that the part of the phage located at the tip of its tail and responsible for bactericidal activity is similar to Colicine. The active unit of Colicine K is estimated to have molecular weight of 60,000 to 90,000.

The loss of individual genetic characters by phage through exposure to ultraviolet radiation, and the restoration of these characters by recombina-

tion within the host bacillus, has been studied by Doermann, Chase & Stahl (81). Their technique permits simultaneous infection of the bacillus by one irradiated phage and several unirradiated T-2 phage particles carrying three mutant gene markers. Provision is also made for the analysis of the phage progeny from individual bacteria. By this technique it has been established that a gene marker originally present in the irradiated phage may be absent from the progeny, even though other markers are present, and that the frequency of such losses is linearly related to the dose. It has also been established that simultaneous loss of two linked markers occurs more frequently than is accountable by chance, and that such double inactivations bear a direct relation to the closeness of the linkage. Unlinked markers are inactivated independently. These observations are considered to be compatible with the hypothesis that ultraviolet damage is localised in the genetic structure, and genetic recombination rescues loci from damage. Interesting questions are raised concerning the replication of radiation inactivated material.

The inactivation of the phages T-1, T-2, T-3, T-5, T-7 and lambda by decay of incorporated ^{32}P has been very fully investigated by Stent & Fuerst (332). It was found that these phages fell into two classes of sensitivity to ^{32}P decay. At the same specific activity of ^{32}P in their DNA, T-2 and T-5 are inactivated three times as rapidly as T-1, T-3, T-7 and lambda. Since the strains of the first class were found to contain about three times as much total phosphorus per phage particle as those of the second, it appears that the fraction of all ^{32}P disintegrations which are lethal is very nearly the same in all the strains. This fraction depends on the temperature at which decay is allowed to proceed and rises from 0.05 at −196°C. to 0.1 at 4°C. and 0.3 at 65°C. Decay of ^{32}P taking place only after the penetration of the DNA of a radioactive phage particle into the interior of the bacterial cell can still prevent the reproduction of the parent phage, though inactivation proceeds at a slightly reduced rate. The T-2 phages inactivated by decay of ^{32}P can be cross-reactivated, i.e., donate some of their genetic characters to the progeny of a mixed infection with no radioactive phage. They do not, however, exhibit any multiplicity reactivation or photoreactivation. A mechanism is proposed whereby the temperature dependence of this type of inactivation can be accounted for by the double stranded structure of the DNA molecule.

Irradiation of oriented tobacco mosaic particles by deuterons *in vacuo* gives a linear relation between inactivation cross section and the sine of the angle between the direction of the deuteron beam and the long axis of the particles, with no influence of angle in a perpendicular plane, in agreement with expectation for a long thin target [Pollard & Whitmore (273)].

Low voltage electron bombardment of T-1 bacteriophage shows that the property concerned with the length of the latent period is situated deep inside the virus, probably at a depth exceeding 100 Å [Davis & Pollard (73)]. This use of very slow electron beams depends on a knowledge of energy-range relations which is unfortunately very inperfect at these low energies. Davis (72) has attempted to determine this relationship experimentally by observing the proportion of a layer of known thickness of either deoxyribonuclease

or invertase which is inactivated as the energy of the electron beam is increased from 100 to 2000 ev. Ranges estimated in this way fall distinctly below Lea's estimates of the integrated range. Consideration of electron scattering might account for some part of the difference.

The extension of the latent period of T-1 phage by exposure to ultraviolet has been studied by Setlow, Robbins & Pollard (315) as a function of ultraviolet wavelength over the range 2300 to 2900 Å at room temperature and at liquid air temperature. A larger amount of ultraviolet energy incident per unit area was needed to produce a given prolongation of the latent period when delivered at low temperature. The action spectrum, expressed as a cross section per incident photon, resembled the absorption spectrum of nucleic acid more closely than did the action spectrum for plaque formation.

Buzzell, Brandon & Lauffer (48) have studied the x-ray inactivation of influenza virus, of virus haemagglutin and of the virus host cell in chick allantoic membrane. The infectivity inactivates exponentially and has a target size equal to about $\frac{1}{8}$th of the virus. The host cells are also inactivated exponentially with respect to support of virus growth, whether or not they were infected at the time of irradiation. The inactivation dose for the host cells is much the same as for the virus itself, namely about 4–5 10^4r.

Comparing three strains of poliomyelitis virus, two strains inactivated exponentially with ultraviolet down to 10^{-6} survivals, but the third inactivated as if it were a mixture of particles of two different sensitivities [Fogh (99)].

The alpha particle inactivation dose of MEF1 poliomyelitis virus has been measured, and a diameter of 16 mμ has been inferred [Pollard & Kraft (272)].

Welsch & Minon (355) report first order inactivation by ultraviolet of strains of actinophage active on actinomycin producing varieties of Streptomyces.

The x-ray and alpha-ray inactivation doses of a variety of virus particles of approximately known size have been measured by Bonet-Maury (32) and interpreted in terms of virus structure.

2. Investigations Employing Physical Variables

In all cases cited above the elementary particles were inactivated with first order kinetics, i.e., the proportion of particles inactivated was linearly related to the logarithm of the dose. In the case of the macromolecules and viruses it is not seriously doubted that the inactivation is, in such cases, brought about by a single ionizing particle. When the same kinetics are observed with larger units of biological organisation, such as bacteria, the same deduction is usually made, but it is less certain because, with the doses generally employed, most irradiated bacilli will have been traversed by more than one electron. This is not the case in the experiments of Marcovich (224, 225). Marcovich describes a procedure whereby the number of *E. Coli* of the lysogenic strain K12(λ) which are induced to develop phage by exposure to ultraviolet or x-rays may be very accurately assayed. By his pro-

cedure interference from spontaneous induction is largely suppressed and it has been shown that the number of bacteria induced is strictly proportional to dose down to doses of less than 1r and independent of dose rate over the range 6–60,000r/min. From the radiobiological standpoint induction behaves like a mutation. Having regard to the size of the bacillus it is clear that an effect which is linearly related to dose down to 1r must be brought about by a single electron, so that independence of dose rate may be assumed for all dose rates lower than those tested, provided the character of the bacterial population which is being induced does not change with time. The system is sufficiently sensitive to respond to natural radiation.

An extremely interesting study of the effects of low doses of alpha-radiation on the movement through mitosis of grasshopper neuroblasts has been described by Rogers (295) . Qualitatively the effects were quite similar to those previously observed with x- and β-radiation but, per unit dose, alpha radiation was the most effective and β-radiation the least effective of the three radiations. When 56 rads of polonium alpha-radiation, delivered by an average of eight particles per cell, were compared with an equal dose of x-radiation, it was found that both radiations either arrested cells in mid-prophase for the period of observation or caused them to revert to earlier stages, but were without effect on late and very late prophase cells. When the alpha-radiation was so reduced as to give an average of 0.8 particles per cell in a one second exposure, a proportion of the cells which were in mid-prophase at the time of irradiation about equal to the proportion not traversed by a single alpha particle appeared unaffected. At this low dose level five recognisable stages of mid-prophase showed different sensitivities to the alpha radiation, the earliest stage being the most sensitive. It was evident that cells in this stage could be arrested by exposure to a single alpha-particle.

(a) *Action spectra.*—

In the case of biological effects produced by ultraviolet irradiation, which certainly do not always follow the same pathway as when produced by ionizing radiation, information as to the initiating step may be obtained by comparing action spectra with absorption spectra [cf. Setlow, Robbins & Pollard (315)].

Raut & Simpson (285) have undertaken a very interesting study of the ultraviolet and x-ray sensitivity of "petite" yeast strains which are characterized by a respiration deficiency, and of the rate of production of this type of cell by irradiation of normal strains. The respiratory deficiency of the petite strain is due to the absence of several enzymes involved in oxygen transport which are normally located on cytoplasmic particles. Genetic tests have shown that petites are not caused by a difference in the nuclear genes, and it has been postulated that they result from loss or inactivation of authonomous self-reproducing non-chromosomal elements. In the case of ultraviolet irradiation the action spectrum for lethality of petites is identical with that of normal cells. When a normal colony is irradiated, as many as 63 per cent of cells, at the 0.05 survival level, may be petites. Since the

action spectrum for the production of petites, as well as that for lethality of both petite and normal strains, is almost identical with the nucleic acid absorption spectrum, the author suggests that energy primarily absorbed in either DNA or RNA is particularly active in both respects. The x-ray lethality curves of normal cells and petites are also identical, but x-radiation is very much less efficient at a given survival level in transforming normal into petite cells. It is concluded that nucleic acid is involved in the inactivation or loss of the cytoplasmic particles and hence that ultraviolet may produce inherited defects which are non-chromosomal in nature.

(b) *Microwave analysis and chemical change.*—

In the case of ionizing radiation we now know a great deal about types of chemical change which might be occurring in the living cells, but we lack information as to those which actually do take place.

Some aspects of the radiation chemistry of solids and aqueous solutions which are of special interest to radiobiology are discussed in relation to each author's own experimental observations in papers presented at the International Radiobiology Conference, Liege, 1954. These include: "Action of Ionizing Radiation on Lipides" by Chevallier & Burg, "The Nature of the Peroxide-like Substances formed in Mice by x-rays" by Horgan & Philpot," "Hydrogen Peroxide Production under Varying Conditions of Irradiation" by Ebert, "Bacteriophage Inactivition under Varying Conditions of Irradiation" by Alper, "After-Effects of Irradiation of DNA" by Butler and Simson, "Physico-Chemical Methods of Protection against Ionizing Radiations" by Alexander & Charlesby, and "Two classes of Protective Agents in the Oxidative Degradation by Gamma Rays of Polystyrene in Chloroform" by Fox.

Although radiation chemistry lies outside the scope of this survey, some mention must be made of the results obtained when living cells and their constituent molecules are examined by microwave spectroscopy. It is evident that the chemists now have to hand a new analytical technique of great power for the investigation of changes which result from the irradiation of organic molecules in the solid state, and the next few years will doubtless see this technique applied to the advancement of our knowledge of processes taking place in the irradiated living cell, despite the great variety of molecules present.

Typical pioneer work in this field is reported in two papers by Gordy, Ard & Shields (120) on the microwave analysis of irradiated amino acids and proteins and of carboxylic hydroxy acids. Molecules were exposed to 50 kv x-rays and examined at 9 kmc and 23 kmc at room temperature. Doses are not stated but may be presumed to have been very large. Most of the molecules examined showed no resonance before irradiation, but almost all did so after irradiation. It is encouraging to find a great diversity in the response of different types of molecule, and some of the observations at once suggest associations in the mind of the radiobiologist, of which many no doubt will prove to be incorrect. Proteins may be very much more stable

than their constituent amino acids. This is exemplified by the particularly interesting case of the cystine-containing proteins of hair, nail, and feather. When these substances are irradiated the resonance observed is attributed predominantly to the cystine, although cystine only comprises 1/10th of the total amino acid content, and it is suggested that when a hole is created by the ejection of an electron, it is quickly filled by an electron from cystine, thereby diminishing the damage induced by the radiation, and possibly even strengthening the polypeptide chain. It was noticed with Teflon [Ard, Shields & Gordy (14)] that resonances which are stable indefinitely *in vacuo* may disappear and give place to others, thought to be due to R—O—O radicals, in the presence of oxygen. Irradiated nucleic acid gave resonances which were stable *in vacuo* and disappeared in the presence of oxygen. Reference will be made elsewhere to the complex problem posed by the protective effect of anoxia, and some of the complexities will be discussed at the biological level. Complexities also exist at the chemical level, and it is to be expected that microwave analysis will contribute to the unravelling of some of these problems.

(c) *High intensity and ultra fractionation.*—

The fact that the early chemical changes following upon ionization and excitation are likely to be transitory has prompted the use of extremely high intensities and ultra fractionation.

Bellamy & Lawton (26) have studied the inactivation of bacteria and of pepsin by 1-mev electrons at dose rates of 140,000r/sec. but have found no essential difference between the inactivation of *E. Coli B*/r at these very high dose rates and at 1000r/min.

Lüning, Lindell & Falk (218) have evaluated the efficiency of extremely high dose rates (2.10^7 r/sec.) in inducing dominant lethals in Drosophila sperm. No differences were observed in hatchability of eggs exposed to 1000r of 370kVp x-radiation delivered in 20 minutes and the same dose of radiation of comparable hardness delivered in 50 μsecs.

Within the last three years considerable attention has geen given to the study of the effects of "ultra fractionated" radiation, in which the dose is delivered in a series of pulses, of from 1–100 m sec. duration followed by pauses of comparable length. In the main, Drosophila eggs, $1\frac{1}{2}$ to $4\frac{1}{2}$ hours old, and Ascaris eggs have been the biological material studied [Dieckmann (80); Hofmann, Kepp & Müller (148); Kepp (171, 172); Müller & Hase (244)]. Currently Hofmann & Müller (149) report a continuation of these studies with Drosophila eggs irradiated by radium β-rays with pulses of from 100 down to 0.26m sec. duration. Sensitivity has been observed to vary with the conditions of pulsing, but the significance and interpretation of these results has yet to be evaluated. The more recent results are summarised by Hofmann & Kepp (146, 147) and by Hofmann (143) who have compared the reactions of skin and of an implanted tumour when irradiated in this way. Apparatus has been designed by Witte (361) to make possible the use of pulses of 2.10^{-7} sec. duration.

(d) *Drying, freezing and low temperature.*—

Using electron irradiation at very high dose rates Bellamy & Lawton (26) (cf. above) have made a number of observations of note concerning the inactivation of different strains of bacteria irradiated in suspension, frozen, or dried. Spores of Bacillus larvae, which are noted for heat resistance, inactivated as a mixed population of which some 20 per cent showed very high radiation resistance. *Streptococcus faecalis*, which was studied as typical of bacteria lacking a complete aerobic respiratory system, nevertheless showed the usual air-nitrogen sensitivity ratio when irradiated in phosphate buffer. This organism withstands freeze drying and, when so dried on a membrane filter, proved very much more sensitive than when irradiated either in liquid phase or frozen. This result was quite unexpected because these bacteria, like most, are more resistant when dried in the mass from phosphate buffer than when irradiated in suspension. In the vaccum dried conditions the sensitivity was the same in nitrogen as in air. The question of sub-lethal damage due to the freeze-drying procedure naturally arises.

Lasnitzki (204) has found that when Ehrlich ascites tumour cells are exposed to 1500r of x-radiation in the frozen state at −79°C. and inoculated into C3H mice, tumours are slower in appearing and grow more slowly. They also show more abnormal division figures in the early divisions after inoculation than material irradiated at room temperature, despite the fact that inocula (15.10^6 cells) of unirradiated frozen cells appear to take as well as fresh material. Essentially similar results were obtained when the frozen cells were irradiated on the bench at a ambient temperature between −79°C. and room temperature. The result was quite unexpected, since many examples are known in which the sensitivity of bacteria and other micro-organisms is depressed by freezing. Slightly greater sensitivity of *B. subtilis* in the frozen state at −78°C. has been reported recently by Proctor *et al.* (281), who point out, however, that this may have been because their bacteria were damaged by freezing. It is not easy to interpret such experiments since there there is much cell death, and presumably, therefore, also many lesser degrees of injury. Some of the difficulties are clearly seen in an analogous investigation by Parkes (256) of the radiosensitivity of ovarian tissue at −79°C. Much is already known of the course of events when unirradiated frozen rat ovaries are grafted, and it was therefore possible to set up several criteria of radiation damage, such as the average latent period, the proportion of takes and the functional life of the graft. At a dose level of 3300r the damage due to irradiation at −79°C. was clearly less marked, in certain respects, than that which resulted when the ovaries which had been similarly prepared by soaking with glycerol-serum were irradiated at room temperature.

3. Cellular Reactions to Nuclear and Cytoplasmic Injuries

Attempts to assign to nucleus and cytoplasm their respective roles in

initiation of radiobiological damage (as distinct from its development and expression) have been continued both by irradiating selected regions of individual cells and by the use of special features of the life cycle of certain types of cell. These questions have been discussed in an informed and stimulating manner by a number of authors in books or review articles which have appeared during the year, notably by Stapleton (325), by Zelle (365) and by Stapleton (326), in "*A Symposium on Radiation Effects on Cells and Bacteria*," by Zelle & Hollaender (366), by Powers (277) by Powers & Ehret (278), by Kimball (176), and by Pomper & Atwood (274).

The beautiful experiments of Zirkle, Bloom & Uretz, in which small regions of individual cells have been exposed to microbeams of protons and ultraviolet radiation, have been commented upon in a previous *Annual Review* [Mortimer & Beam (238)]. Two further reports have appeared during this year (31a, 368). The cells are followed after irradiation by time lapse cinematography, and some interesting examples of the microscopical appearances of the irradiated cells are given and discussed in review articles by these workers. The experiments provide very direct evidence that to destroy the function of the centromere or to produce chromosome breaks it is necessary that the protons should pass either through or in the immediate vicinity of the structure which is being damaged. On the other hand, ultraviolet irradiation of cytoplasm may destroy the spindle and derange metaphase. Spindle effects were only observed with protons when extremely high doses were given to the cell.

By exposing to x-rays either the anterior or posterior half of Drosophila eggs prior to first cleavage, Ulrich (341) has established both a difference in the shape of the mortality curve and a 180:1 difference in the 50 per cent survival doses. The survival is exponentially related to dose in the former case and sigmoidal in the latter. Thus, in Drosophila as in Habrabracon, cytoplasmic injury alone, induced by moderate irradiation prior to cleavage, is relatively unimportant to the development of the embryo. Lesions induced by exposure to low doses of radiation sufficient to depress survival to the 10 to 50 per cent level only develop when the nucleus has been directly irradiated.

Taking advantage of the asymmetric location of the nucleus in Habrabracon eggs prior to second meiotic metaphase, Von Borstel & Wolff (351) have been able to compare hatchability of eggs after exposure to ultraviolet when the nucleus was irradiated and when it was excluded from the field of irradiation. At moderate dose levels survival is critically dependent on irradiation of the nucleus. The experiments have shown that survival is exponentially related to dose when the nucleus is irradiated, and sigmoidally related when only cytoplasm is irradiated, and further that by the criterion of hatchability, photo-reactivable injury is related to events within, limited to, and governed by, the egg nucleus.

Androgenesis, i.e., the development of a fertilized egg without participa-

tion of its own nucleus, which is exhibited by irradiated eggs of the parasitic wasp Habrabracon, is a powerful tool in the hands of Whiting (357) for the investigation of the relative importance of nuclear and cytoplasmic injuries in the development of irradiated eggs. When eggs are irradiated at Metaphase I, androgenic males first appear at a dose level of 2400r, which is a measure of the sensitivity of the egg nucleus to lethal damage. Androgenic males continue to appear as the dose is raised, and only cease at 54,000r. Thus a normal haploid embryo may develop from an unirradiated nucleus in cytoplasm exposed to 22 times the dose which is lethal to the nucleus.

Noting that haploid Habrabracon embryos derived from unfertilized eggs are more resistant to both x-rays and nitrogen mustard than diploid embryos derived from fertilized eggs when both are treated at the cleavage stage of development, Clark & Beiser (56) have proposed this test as a method of screening cell poisons as to their mode of action, and applied it to some amines, sodium azide, and potassium cyanide.

The inactivation of haploid yeast is exponential for x-rays and sigmoid for ultraviolet radiation. The inactivation of dipoloid yeast is sigmoid for both radiations. A study of the additivity of x-rays and ultraviolet [Uretz (342)] has shown that the two radiations exhibit a great degree of interaction in the diploid, but almost no interaction in the haploid. These results accord well with the view which is now gaining general acceptance that the haploid organism offers a unique set of sites for inactivation, while these are replicated in the diploid. Each site may be inactivated by a single electron, but not by the absorption of a single ultraviolet photon. Since successive photon absorptions may be separated in time by as much as one and one half hours without loss of effectiveness, they probably represent different electron excitations, all of which are necessary for the inactivation of the site. It is evident that in the haploid organism, a site in which some, but not the required number, of photon excitations, has occurred is appreciably more apt to be inactivated by x-rays. It is also concluded that to an appreciable extent the sites involved in x-ray and ultraviolet inactivation are the same. Further light is thrown on the mode of inactivation of haploid and diploid yeast by the work of Mortimer (237) using haploids of opposite mating type which will conjugate under appropriate conditions. The inactivation of haploid yeast is satisfactorily interpreted in terms of the induction of recessive lethal mutations. The survival of zygotes is found to be the same after the irradiation of either parent cell, but is very much greater than when both parents are irradiated or when the diploid cell is irradiated, and it is concluded that recessive lethal mutations only account for about 1/15th of the diploid cells which fail to survive. When diploid cells or cells of higher ploidy are irradiated, failure to survive is ascribed mainly to the induction of dominant lethal damage.

Maisin, Lambert & van Duyse (222) make a preliminary announcement of the production, by repeated irradiations, of a resistant strain of yeast. The

radioresistant character appears to be stable, but whether it has arisen by selection or mutation has not yet been determined.

James (161) brings forward evidence that exposure of vegetative diploid yeast cells to ultraviolet may induce a reassortment of genetic loci. Homozygous lines are obtained from the heterozygote which are believed to be due to segregation rather than mutations, since complementary dominant and recessive homozygous lines are often obtained. Moreover, haploids rarely show change from dominant to recessive, and irradiated homozygous recessive cells only rarely show change to the heterozygous state. The phenomenon is not rare since, for a particular allelic pair, it may be seen in 2 per cent of survivors at the 50 per cent survival dose level. In a second paper [James & Lee-Whiting (162)] further evidence is presented which indicates that the segregation arises from mitotic crossing-over.

Damage to cytoplasm, especially at high dose levels, may be a factor influencing the development of nuclear damage, even if not lethal in itself. Some experimental evidence on this point has been provided by studies of induced mutation frequencies in moulds. Mutational frequency is rarely linearly related to ultraviolet dose except over a very limited range of low doses. The proportion of mutants among survivors from ultraviolet irradiation generally reaches a peak and then declines as dose is increased. x-ray induced mutants on the other hand generally increase linearly over the whole range investigated, but an exception was observed in one of the Actinomycetes by Kelner (170) in 1948. This has been followed up by Newcombe (248) who has made a detailed study of the influence of physiological conditions on x-ray induced mutations in streptomyces. Exceptional sensitivity to x-ray induced mutations was observed to extend over the whole binucleate stage and fractionation experiments indicated that nuclear division brought no release from the earlier saturation of the capacity for mutation inducation. It is suggested that the nonlinear dose-response relationship is in fact due to physiological control on mutability of the genetic material. Latarjet (205), in discussion of this paper, adds a striking example of a steep fall in ultraviolet mutation frequency of phage T-2 exposed within the host bacillus at a critical ultraviolet dose level.

Unicellular organisms, by peculiarities in their mode of life and reproduction, provide special opportunities for the study of some radiobiological problems, despite the rather great radioresistance of these organisms when reproducing vegetatively. The whole field is reviewed authoritatively by Kimball (176). One very interesting situation is presented by autogomy in Paramecium which uncovers genetic damage to the micronucleus. Mitchison (234) has studied the effects of ultraviolet on Paramecium aurelia in order to test Raffel's hypothesis (284) that mortality at autogomy in lines in which autogomy has been delayed is due to lethal mutations in the micronuclei. He finds the simple hypothesis unacceptable. Either the simple hypothesis must be extended, or damage to the cytoplasm or macronucleus,

or both, must be postulated to account for his observations. In view of the selective absorption of ultraviolet and the distributed absorption of x-ray energy within the cell, Mitchison's conclusions do not, of course, necessarily apply to x-ray damage; indeed, some effects observed by Mitchison with ultraviolet were not seen by Kimball using x-rays.

4. Investigations Concerned with Cell Metabolism

Cell metabolism and cell structure are no doubt intricately and reciprocally connected so that what is seen in the irradiated cell is a downward progression of both structure and functional metabolic activity. It has hitherto proved much easier to detect changes in structure than changes in metabolism at early times after exposure to low doses of radiation, and we may therefore consider first those investigations which hold out some promise of giving an indication of some rather immediate interference with metabolism.

(a) *General metabolism.*—

Gordon (119, 119a) reports in some detail upon a metabolic disturbance seen in plant tissues immediately after very low doses of radiation, of which there are still extremely few examples which have been credibly reported. The effect concerns the synthesis of auxin by the irradiated kidney bean plant. It was shown by Skoog (319) over 20 years ago, in what must be one of the earliest papers on the radiation chemistry of enzymes in aqueous solution, that auxin, like other enzymes, is readily inactivated at sufficiently high dilution and more readily in the presence of oxygen. It was also thought that auxin was inactivated in the oat coleoptile, but the work of Gordon, as well as that of Riley, failed to confirm this at low doses. Inactivation only occurs after doses in the neighbourhood of 30,000r. Oat coleoptiles exposed to such doses, exhibited a reduced rate of cell elongation which may be offset by addition of auxin after irradiation. When estimated quantitatively it was apparent that auxin in the coleoptile is neither specially radiosensitive nor preferentially inactivated. Auxin is likewise not inactivated by breis of irradiated plants. When, however, whole seedlings of kidney bean or cocklebur are exposed to doses of as little as 25r significantly less auxin is extractable immediately after irradiation than from control plants. After doses of up to ~1000r the auxin level returns approximately to normal in two weeks. Auxin synthesis is known to occur in meristematic tissue and the time course of auxin depression was typical of that for other aspects of meristematic activity. The association with meristematic activity was confirmed by irradiation of apical buds. The metabolic aspects have been worked out by study of the ability of irradiated seedlings and of cell free homogenates irradiated *in vitro* to synthesize auxin from tryptophane. After doses ~100r to the seedling, less auxin is formed from infiltrated tryptophane, and its precursor, indoleacetaldehyde, accumulates, indicating the enzyme responsible for the final step in auxin synthesis as the site of radiation damage. Similar doses inhibit the conversion of the aldehyde to

auxin by cell free homogenates of coleoptiles. The enzyme responsible for the conversion was tracked by ultracentrifugation to a soluble fraction of the cytoplasm heterogeneous with respect to molecular weight and estimated to be present in about 10^{12}–10^{13} molecules per gram tissue. Since this is inactivated by x-rays at an initial rate of about 10 per cent per r, i.e. 10 per cent per 2.10^{12} ions/gram, an apparent ionic efficiency of 10^{-1}–10^{-2} was derived. This is an extraordinarily high value for an enzyme irradiated in the presence of a large bulk of competing material, and leaves unexplained the channeling of energy into this particular enzyme, if an explanation is sought in terms of an immediate inactivation of the enzyme itself.

In the course of a study of the effects of x-radiation on isolated plastids, irradiated *in vitro*, Sissakian (318) and his co-workers have observed that a dose of 5000r affects the absorption and elution of enzymes by the chloroplasts and leucoplasts of sugar beet, as well as the activity of the enzymes themselves. Vacuum infiltration has been used to study the synthesis and breakdown of saccharose and the biosynthesis of protein in rye seedlings, processes which were affected during the first day at the 15–30,000r dose level. In the same seedlings nucleic acid synthesis was followed, after doses of 5000r and upwards, DNA synthesis being in general depressed, while RNA accumulates. This was associated with a fall in the ribonuclease activity. In this brief survey paper no indications are given of associated histological changes in the seedings.

Rosenfeld *et al.* (299) appear to have broken new ground from the radiobiological standpoint in demonstrating an effect of a moderate dose (2000r) of radiation on the synthetic activity of the isolated adrenal. The results are the more interesting because the effect was evident during the course of a 1 to 3 hour irradiation and in an organ in which histological damage is minimal or absent at the dose levels employed, so that there does not appear to be a *prima facie* case for suspecting the effects to be secondary to cell degeneration or changing cell population. The experiment consisted in irradiating continuously with Cobalt-60 γ-rays pairs of isolated calf adrenal glands which were being perfused with calf blood (previously freed from vasoconstrictors and interfering constituents by passage through rat liver) at 1000 ml/hr. In the first experiment steroid precursors were added to the blood passing through one of each pair of adrenals and in the other experiment ACTH was added to the blood passing through one of the adrenals to stimulate steroid output. Hourly collections of effluent blood from the irradiated adrenal showed substantial decreases in synthesis of steroids or steroid output under ACTH stimulation compared with controls. In a later experiment not involving the use of irradiation, Rosenfeld (298) compared the output from ACTH stimulated adrenals perfused with blood and with artificial media. He observed that the steroid output was remarkably independent of a variety of added substances, including potassium and histamine, but that oxygen was essential, ATP increased output, and the thresh-

old for ACTH stimulation was lower if the red blood corpuscles were present in the perfusing fluid. It is therefore of some importance to exclude the possibility that in the irradiation experiments the gland was responding to minute radiation-induced changes in the plasma or corpuscles of the perfusing blood, and this does not yet appear to have been done.

Although the subject of radiation chemistry is outside the scope of this survey, reference may be made to a number of papers in which the inactivation of an enzyme or other biologically important molecule has been studied, particularly when the inactivation of the molecule as it occurs within the cell has been compared with that of the same molecule in isolation or in aqueous solution. These include a study of the inactivation of dried bacterial α amylase as a function of LET [Jagger & Hutchinson (159)] and of β amylase by deuterons in relatively pure form, in an immediately extracted form, and within 25μ sections of barley seeds which were vacuum dried. The β amylase responded to ionizing radiation in essentially the same manner under all three conditions [Jagger & Wilson (160)]. The deuteron and electron inactivation of invertase within vacuum dried yeast cells was found to be the same as that of a commercial preparation, and the deuteron inactivation of the cytochrome oxidase and succinic dehydrogenase of dried lysates of *B. subtilis* was the same as that of the enzyme within dried cells. Thus, under these somewhat artificial conditions, with water absent and no metabolism, which were chosen in order to isolate for study damage due to direct ionization of the organic structure of the cell, the inactivation of the enzymes within the cell and outside the cell was indistinguishable. Moreover, 140,000r of x-rays to *B. subtilis* and 200,000r to barley in aqueous suspensions at 4°C. were without influence on the activity of the extractable enzymes [Powell & Pollard (276)]. However, it has been observed that hyaluronidase, when dried *in vacuo* together with hyaluronic acid, is more sensitive to deuterons than when dried alone [Setlow & Doyle (314)]. It appeared that an ionization in the substrate could inactivate the enzyme.

The lens offers certain special advantages for the study of metabolic changes resulting from exposure to ionizing radiation. On the technical side inter-animal variability may be eliminated by using the other lens as a control. These studies are basically interesting in that they concern an organ which eventually shows cytological damage and changes in metabolic pattern after comparatively small doses of radiation. A systematic examination [Pirie, van Heyningen & Boag (265); van Heyningen, Pirie & Boag (347)] of many aspects of lens metabolism has, however, consistently failed to reveal any radiation induced changes within the first 24 hours after irradiation. Currently Pirie, van Heyningen & Flanders (266) report a detectable fall in glutathione at 24 hours after exposure of the lens of very young rabbits to 1000r, but no change was detected in high energy phosphate groups of ATP and ADP or in hexose monophosphates, hexose diphosphate, phospho-glycerate or inorganic phosphate within a few days of irradiation.

The ATP-ase activity of liver and muscle homogenates, irradiated *in vitro* at dilutions of 1:10 to 1:20,000 and doses of 5 to 100kr, have been measured by Hornykiewytsch & Stender (156). Under optimum conditions of maximum dose and maximum dilution, activity is reduced to 5 per cent, but the effectiveness of a given irradiation diminishes the more concentrated the preparation, and it is concluded that a direct effect on ATP-ase is probably not important in the irradiation of the living cell.

Hagen (133) has looked for a radiation induced oxidation of reduced cytochrome in living yeast and *E. coli* cells when irradiated in the absence of oxygen, but found no evidence of such an effect after doses of up to 30,000r though precautions were taken to avoid the reversal by metabolic processes of any oxidation that might have been produced.

Langendorff & Hagen (201) using the same procedures, first showed an oxidation of reduced cytochrome in *E. Coli* at 45,000r, which is nearly two orders of magnitude greater than the dose of 1000r which suffices to reduce to 50 per cent the number of organisms capable of forming a colony.

Since Potter & Bethel (275), and Ashwell & Hickman (15) have reported that the oxidative phosphorylation systems in spleen and thymus are affected following irradiation *in vivo*, and effects have been detected as early as 2 hours after irradiation [van Bekkum (343); van Bekkum and co-workers (344), van Bekkum & Vos (345)], Ord & Stocken (254) have looked for effects of radiation on oxidative phosphorylation and ATP-ase activity of rat spleen irradiated *in vitro*. Doses of up to 960r delivered in 12 minutes failed to produce any significant changes. Lymphocytes degenerate so rapidly after whole body exposure of an animal to moderate doses of radiation that the spleen and thymus are evidently not the most favourable organs in which to seek the influence of irradiation on metabolism—more especially as humoral factors may contribute largely to the cell degeneration. Petersen, Fitch & du Bois (264) have clearly shown that several forms of radiobiological damage manifested by the rat spleen after whole body irradiation are, by comparison, negligible after an equal dose to the exteriorised spleen. Criteria included increase in ATP-ase and 5 nucleosidase activity per gram of spleen, decrease in citrate synthesis, DNA per gram, and histological damage. As regards histological damage 800r to the exteriorised spleen approximately matched 100r to the whole animal [cf. also ref. (169)]. Since first observations were made at six hours it is perhaps arguable, or even probable, that the whole of the observed changes are referable to changes in cell populations resulting from onset of degeneration, which is very rapid in the spleen. It may be, therefore, that the enzyme activities of the more sensitive elements are not being measured, but the results still demonstrate that interpretation of both cytological disturbance and metabolic changes evaluated per gram of tissue in totally irradiated animals, is equivocal from the standpoint of cellular radiobiology.

Simonnet, Demaux & Kahn (317) have observed differences between damage to rat spleen after whole body irradiation and irradiation of the

exteriorised spleen in exactly the same sense. Since the smallest *in vivo* dose was 1000r, the quantitative differences were not quite so great as observed by Petersen, Fitch & du Bois (264) [c.f. also Hoffmann & Matscke (150) and Falk (96)].

The experiments of Booz (34) appear at first sight to be at variance with this conclusion. Following the work of Booz, Betz & Firket (35), who had found that spleens from animals exposed to upwards of 10,000r were able to regenerate completely when grafted into an unirradiated animal, Booz has grafted spleens which had been irradiated *in vitro* with doses of 10,000 to 60,000r and obtained regeneration essentially comparable to that obtained when the spleens were irradiated *in vivo*, i.e., degeneration, followed by complete regeneration after 10,000r, and failure to regenerate only after 60,000r. The evaluation of this type of experiment obviously hinges on whether irradiated cells give rise to the new splenic elements, or merely constitute a framework within which the cells of the unirradiated animal proliferate to repopulate the organ. This question does not yet appear to have been settled in the case of the spleen. Regeneration of haemapoetic elements in irradiated animals by proliferation of transplanted marrow has now been firmly established [Ford *et al.* (101)]; Barnes & Loutit (21, 22), Lindsley, Odell & Tausche (212); Main & Prehn (221)].

In vitro exposure to 150,000r of x-radiation does not measurably change the rate of leakage of para-aminohippuric acid out of slices of rabbit kidney. Suggestions are made regarding the effects which have been observed *in vivo* [Mendelsohn (228)]. Effects of x-rays on sugars in barley seeds have been studied by Ehrenberg & Jaarma (89).

The very great stability of cell metabolism with respect to heavy irradiation is not without its advantages. It is well known that very great difficulty is experienced in growing clones of mammalian cells in culture medium from a single cell or from a few cells. A condition of success is the conservation of diffusible cellular products, which is technically difficult when only a small number of cells are present initially. Puck & Marcus (283) have solved this problem very beautifully by taking advantage of the fact that radiation inhibits cell reproduction at dose levels which leave many metabolic functions of the cell intact for a considerable time. A large number of fairly heavily irradiated cells may thus serve as "feeder" cells to nourish the cells of interest until they are sufficiently numerous to be self-sustaining.

Morczek (236) has studied the movement of potassium outwards and of the compensatory movement of sodium inwards in irradiated heparinised human red blood corpuscles, and finds effects which increase with the time after irradiation and which, during the first three hours after irradiation, increase linearly with dose up to 16,000r.

Meissel (227) summarises radiobiological studies by himself and coworkers with micro-organisms. The x-ray doses which were used generally go up to 10^6r, and are appropriately delivered by a tube (dissipating 60 kw at 9 ma current) giving 21,000r/min. at 6 cm. Data are presented for yeast

showing increases in amino groups, —SH groups and free methionine, release of phosphate and of nucleotides from the cell, and various aspects of phosphorus, nucleic acid, steroid and vitamin metabolism. Interference with steroid metabolism is emphasised as an effect observed after much lower doses (2 kr) than most other metabolic disturbances. Morphological changes are illustrated by phase contrast photomicrographs; the emphasis is on direct rather than indirect action of radiation, and on morphological and physiological changes, including disturbance in nucleic acid, fat, and steroid metabolism, rather than chromosomal and genetic damage. In both cases the emphasis may be related to the high dose range investigated.

Morczek & Mücke (235) have studied the Q_{O_2} of irradiated *Allescheria boydii*, 1699, an ascomycete which requires biotin. Relative to controls the respiration was slightly more depressed in the presence of biotin, but in both cases doses of at least 100kr were needed to have an immediately detectable effect on Q_{O_2}. The largest doses did not reduce Q_{O_2} below 40 per cent of control, but this "residual respiration" was completely eliminated by heavy metals such as $HgCl_2$ and Ag_2SO_4 at 10^{-3}M.

Nizet & Herve (249) report a stimulation of the rate of *in vitro* synthesis of haemaglobin by reticulocytes of blood taken from dogs immediately after exposure to 500r. On occasion, stimulation was observed simply by irradiating the blood *in vitro* or by mixing it with plasma from an irradiated dog. The experiments were evidently carried out with great care. The synthesis of the haem and the globin were separately estimated by the rate of incorporation of suitable C^{14} labelled precursors. However, the number of dogs studied so far is small and the results rather variable, so that more data are desirable in order to assess the significance of these results. A decrease in uptake of iron by duck erythrocytes *in vitro* following whole body x-irradiation has been observed by Klein (186).

Studies of respiration and anaerobic glycolysis of skin have shown evidence of impairment which changes with the time after irradiation. Distinct differences between Q_{O_2} of control and irradiated skin were measurable within the first hours after exposure to 12,000r [Gahlen & Stüttgen (103)].

(b) *Nucleic acid metabolism.*—

The search for evidence that irradiation of the living cell damages particular cell constituents or particular metabolic processes is reviewed in some detail by Errera (95) who concludes that the system which is most evidently disturbed is that concerned with DNA synthesis, but points out that this may not be because the system is more sensitive, but simply that it exercises a more dominant role.

A number of authors report on the influence of x-radiation on DNA metabolism. Thus, Pelc & Howard (261) have used the autoradiograph technique to study the proportion of *Vicia* meristem cells examined in interphase at a chosen time (12 hours) after different doses of x-radiation in which ^{32}P has been built into DNA in a form not removed by acid hydrol-

ysis. This proportion is about 40 per cent of controls for all doses examined between 50r and 200r. This result is interpreted to be due to a greater radiosensitivity to delay or inhibition of DNA synthesis on the part of cells which are in approximately one third of the cell cycle at the time of irradiation than in cells at other stages of the cycle. It is suggested that many of the results reported in the literature in which the amount of tracer incorporated into DNA at early times after moderate doses of radiation had been observed biochemically to be roughly half that in controls could be explained on the hypothesis that only a part of the irradiated cell population is primarily affected. Kelly *et al.* (169), have studied the incorporation of ^{32}P into the DNA of many organs and of tumours of the mouse during a two to four hour period at intervals after doses from 300 to 2500r. In every case a fall in specific activity of DNA phosphorus was observed shortly after irradiation. It is of interest that despite the fact that the mammary carcinoma and the lymphosarcoma examined in these studies had about the same growth rate, mitotic index and short term incorporation of P^{32} before irradiation, they responded quite differently to irradiation. The four hours incorporation of ^{32}P fell to about 50 per cent in the radioresistant carcinoma and remained at this level for six days, whereas in the radiosensitive sarcoma it fell precipitately but was above the control level from the third to the sixth day after irradiation. The mice were totally irradiated and the results are interpreted with great caution by the authors, who point out that, in almost all cases, the observed changes in specific activity could be accounted for on the basis of changes in cell population. This does not rule out the possibility of a specific inhibition of DNA synthesis, but suggests that experiments with other materials may be better suited to the investigation of this problem.

The importance of the whole body contribution to the influence of radiation on nucleic acid levels in the thymus is clearly demonstrated, for example, by the work of Weymouth, and co-workers (356) who observed an entirely different pattern in the time variation of nucleic acid per cell in the thymus in the case of totally irradiated animals and animals having one limb shielded. In the light of what is now known of the repopulation of bone marrow from unirradiated regions, it is likely that the observed effects were related to repopulation of the thymus, and do not reflect a change in DNA metabolism of irradiated cells.

In the experiments of Holmes & Mee (155) on the effect of irradiation on DNA synthesis and mitotic activity in regenerating rat liver, localised irradiation is used, so that secondary effects arising from the irradiation of bone marrow and spleen are minimised. Several points clearly emerge. DNA synthesis as judged either by incorporation of ^{32}P or glycine-^{14}C is very much more readily disturbed if the cells are irradiated shortly before the onset of synthesis (12 hours after hepatectomy) than if irradiated after synthesis has commenced. Measurable disturbance is induced by 150r delivered prior to the onset of synthesis, but only after a lapse of several hours. Moreover, the disturbance is of the nature of a temporary arrest rather than

a permanent inhibition, since 14 hours after irradiation synthesis reaches as high a level as it was a few hours previously in controls. After 450r delivered prior to the onset of synthesis the delay has increased to about 14 hours, but the peak level of specific activity is as high as in controls. It may be significant that after this treatment almost every cell coming into mitosis shows chromosome structural damage. At least 2000r are needed to affect DNA synthesis if delivered after the beginning of the synthetic period, but the effect is immediate and probably irreversible. A much smaller dose delivered during synthesis, though without effect on the synthetic process itself, suffices to delay the onset of mitosis, thus confirming that the latter effect of radiation is not conditioned by interference with DNA synthesis.

The effects of γ-radiation and of aminopterin on the metabolism of Formate-^{14}C, glycine 2-^{14}C and Adenine 8-^{14}C by chick embryos suspended in saline has been studied by Passonneau & Totter (257), more especially the incorporation of these compounds into RNA, DNA and protein during a two hour period immediately following treatment. It has been observed that the incorporation of formate, but not that of the other compounds, is sensitive to aminopterin administered in concentrations as low as one microgram per ml. The γ-radiation was only effective at doses above 1000r, and it was concluded that at this dose level γ-radiation does not have a direct and immediate effect on DNA synthesis in the 60 hour chick embryo.

In essential agreement with the observations of Klein & Forssberg (185) on the influence of radiation on the growth and metabolism of the Ehrlich ascites tumour, Gardella & Lichter (105) find that when the cells of the Yoshida rat sarcoma are exposed in the peritoneal cavity to 1000r (the whole animal receiving this dose), they steadily enlarge over a 48 hour period without synthesizing DNA, so that over the period 24 to 48 hours volume, water content and total nitrogen, have roughly doubled, while the DNA per cell is unchanged.

Lajtha, Oliver & Ellis (196) and Lajtha & Suit (197) report on the effects of x-radiation on the synthesis of DNA in bone marrow cells growing in culture medium. By autoradiographic methods it is found that synthesis occurs in the middle of interphase, and the timing in some respects resembles that in root tip meristems. Observations with labelled phosphate, adenine, and formate, gave concordant results. 5000r of x-radiation had a marked effect on DNA synthesis and it was concluded that cells in the synthetic period are immediately and completely prevented from synthesizing DNA by this dose of radiation.

The effects of single doses of x-rays delivered locally or as whole body irradiation on the DNA of bone marrow have been studied by Rodesch *et al.* (294).

The appearance of Feulgen-negative nuclei in Habrabracon embryos developed from eggs treated in meiotic metaphase I with lethal doses of x-rays or nitrogen mustard has been observed by Von Borstel (349) and is considered to arise from an interference with DNA synthesis.

The amount and specific activity of the DNA of thymus and spleen of Wistar rats which had received toxic doses of ^{32}P has been examined by Harrington & Lavik (135). A dose of 4.5μC per gram body weight was estimated to deliver 500r to the thymus in 48 hours. Striking decreases in the DNA per gram of tissue in the thymus and spleen and smaller decreases of RNA in the thymus were observed. Since, however, the DNA per nucleus of the thymus was unaltered, it was concluded that the fall in DNA was due to the loss of DNA rich cells. The lower specific activity of DNA and RNA was ascribed to inhibition either of synthesis or of new cell formation.

It seems clear that DNA synthesis may be arrested by doses of radiation comparable with those which produce cytological damage in the most sensitive cells, provided the cells are irradiated berfore the onset of synthesis. DNA synthesis appears equally affected whether judged by the incorporation of phosphorus, or of one of the purines or pyrimidines, by contrast with, for example, aminopterin which also interferes with DNA synthesis but appears to act at the level of thymine synthesis. While comparable doses are required, in some cells, to inhibit DNA synthesis, to produce chromosome aberrations and to delay the onset of mitosis, interference with DNA synthesis is certainly not the sole cause of radiation damage to chromosomes, since such damage may be caused by irradiating cells in which DNA synthesis is complete, and it equally cannot be the reason why cells in late interphase or prophase are temporarily arrested from entering division. In the root tip and in the regenerating liver the two processes are seen to be quite distinct in regard to radiation damage.

It may be that some of the reversible effects of radiation are more concerned with the colloidal properties of nucleoproteins than with the nucleic acid and protein individually. This is suggested by the following experiments of Kaufmann and his colleagues, as well as by unpublished experiments of Anderson. The appearance of abnormally large cells, or cells with swollen nuclei and nucleoli is a striking feature of many different kinds of irradiated cell populations. When seen at a time after irradiation comparable with, or greater than, the intermitotic time for the cells in question, it may be an expression of the fact that radiation in general interferes with reproduction and cell division at a much lower dose level than that at which it interferes with the synthesis of protein and other basic constituents, but when seen within a short time of irradiation swelling clearly reflects some alteration in the colloidal properties of cellular materials. Kaufmann, McDonald & Bernstein (168) have performed a number of interesting experiments on this latter type of swelling, both by studying the ability of salivary gland cells of *Drosophila* which had been irradiated in the larva to swell when subsequently fixed and treated enzymatically, and by measuring the effect of irradiation on the viscosity of calf thymus jells. In the former case irradiation reduces the capacity for swelling, and in the latter it reduces the high viscosity of the native material. Two points of special interest emerge. Effects in the salivary gland cells are observed after only 250r, are immedi-

ate, but somewhat more pronounced a few hours after irradiation, and have disappeared at 24 hours, so that the time pattern and dose pattern is quite similar to that for mitotic inhibition or chomosome stickiness in a sensitive type of cell. In experiments with calf thymus nuclei, and in others with an artificial nucleoprotein, the dose range studied is also 250 to 1000r and in this case experimental evidence is adduced in support of the authors' view that under conditions which prevail in the swollen cells, nucleoprotein is much more affected by irradiation than protein-free DNA. It appears to be well established that in regenerating liver cells [Holmes & Mee (155)] and *Vicia* root tip cells [Howard & Pelc (157)] mitotic inhibition cannot be dependent on failure in DNA synthesis, but there naturally comes to mind the question as to whether the phase at which cells are particularly easily arrested at the onset of mitosis is one which is critical in relation to the formation of new associations between DNA and protein.

5. Some Questions Relating to the Production of Chromosome Aberrations

The conflicting views which exist regarding the influence of dissolved oxygen on the production of chromosome aberrations will be considered in later sections.

Further experiments by Sax, King & Luippold (307) have not entirely resolved the discrepancies between the observations of different authors regarding the effects of fractionated x-ray doses on the frequency of chromatid and chromosome aberrations induced in Tradescantia microspores. Attention was concentrated on chromosome aberrations, since mitotic delay induced by the irradiation should be without significant influence on the aberration frequencies seen in metaphase four days later. The chromatid aberrations were also studied. Temperature and illumination were controlled. Four fractionated doses of 30r with rest intervals varying from five minutes to eight hours were compared with a single dose of 30r and a single dose of 120r. Two-hit chromatid aberrations decreased, with a half period of about five minutes, to the frequency expected for four completely independent exposures. Chromosome aberrations fell with a half period of about an hour, but to a level lower than expected, indicating a small depressing effect of radiation on sensitivity to subsequent chromosomal damage such as was postulated by Lane (200). Isochromatid breaks did not behave entirely in accordance with expectation for a one-hit type of aberration. It is evident that when 2-hit aberrations are interpreted in accordance with the classical theory of Sax, elaborated by Lea & Catcheside, the duration for which breaks remain open depends on the stage of nuclear activity, and may vary from five minutes for *Tradescantia chromatids* to one hour for *Tradescantia* chromosomes, several hours for *Vicia* seed chromosomes [Wolff & Atwood (362)] and many days in *Drosophila* sperm [Kaufmann (167)]. The validity of the classical theory has, however, been questioned by Revell (288). On the basis of a study of chromatid aberrations induced in *Vicia* root tip

meristems by the radiomimetic chemical di(2:3 epoxypropyl) ether Revell has abandoned the generally accepted view that chromatid exchanges arise by illegitimate fusion between broken ends of chromatids, in favour of the hypothesis that the exchange is the primary event, and that breaks result from the incompleteness of a proportion of exchanges. On this view the isochromatid break is a special class of exchange. The frequency of exchanges was observed to increase approximately in proportion to the concentration of diepoxide to which the roots were exposed for a given time. These exchanges were extremely nonuniformly distributed along the length of the chromosomes, being observed to occur predominatly between heterochromatic regions. In these and some other respects the chemically induced aberrations appear to differ from those induced by ionizing radiation, and might be better fitted by the "contact first" type of hypothesis. These observations are of great interest, but in the view of the writer do not in themselves appear to constitute a sufficient ground for abandoning the classical view of radiation induced chromosome exchange [Gray (125)]. Revell, however, considers that his own observations on x-ray induced aberrations are better interpreted in terms of the exchange as the primary event. The fact that interchanges produced by exposure of *Tradescantia microspores* and *Vicia* meristem cells to x-rays increases as the square of the dose is not so naturally accommodated by the new hypothesis as the old. It may be noted, however, that there are classes of interchange, notably those produced by irradiation of dormant barley seeds, which Caldecott (49) observes to be produced with a frequency which is linearly related to dose.

Bora (37) has made a very thorough study of the aberrations produced in *Tradescantia bracteata* by fractionated x-irradiation. Doses of from 180 to 360r were given as single exposures or as two equal fractions separated by intervals of one to thirty-two hours. A variety of dose rates were employed, and while plants were always at 25°C. when irradiated, they were held during the interval between irradiations at 25°C. in one series and 10°C. in another. The frequencies of those types of aberrations which were influenced by fractionation fell steadily with increasing interval between the two fractions under all conditions investigated, and showed no evidence of returning to higher values with prolongation of the interval up to 32 hours. The observations were considered to confirm the previous conclusion that most chromosome exchanges take place within a short time of irradiation. In addition to its main purpose the paper gives information about the influence on aberration frequency of the stage of development at the time of irradiation, on the influence of temperature on radiation development, and of dose rate on rate of entry of the cells into division.

When *Tradescantia paludosa* roots, grown from cuttings in culture solution, are exposed continuously for 24 or 48 hours to gamma radiation, the proportion of anaphase figures which are abnormal by the end of the irradiation increases with dose rate and appears almost to reach an equilibrium value in the course of 24 hours, as might be expected on account of cell

turnover in the meristem. At 100r per day some 60 per cent of the anaphases are abnormal after 24 hours, and 76 per cent after 48 hours [Mikaelsen (230, 231)].

Following up Sparrow's observation of the extremely high sensitivity of meiotic metaphase-I cells in Trillium to the induction by x-rays of achromatic lesions which appear as visible fragmentation at the microspore division, Deschner (76), Deschner & Sparrow (77), have compared aberrations induced by exposure of cells in various stages of meiosis to x-rays and to slow neutrons, looking particularly for a possible relation between breaking and rejoining which might account for the varying proportions of fragments and of bridges and dicentrics. No direct relationship between fragmentation and rejoining was found. The ratio of the two types of aberration varied with the developmental stage irradiated in the same general way for both radiations. Metaphase was most sensitive to fragmentation by neutrons as well as x-rays. The influence of stage on radiosensitivity was, however, much less with the neutrons than with x-rays, just as in barley the neutron induced lethality is less dependent than x-ray induced lethality on soaking.

The induction by x-radiation of half chromatid exchanges, described by Swanson (335) and Crouse (68) has been investigated in *Tradescantia* by Sax & King (306) with special reference to the time of chromosome doubling and the timing of DNA synthesis. As regards x-ray induced aberrations, *T. microspore* chromosomes react as single threads when irradiated during the resting stage, as double threads when rayed at prophase and four-partite threads when rayed at pro-metaphase and metaphase. These sub-chromatid breaks are also described by Kihlman (175) in an important paper mainly concerned with a detailed comparison between aberrations induced in allium root tip cells by x-rays and by the very closely radiomimetic chemical 8-ethoxy-caffeine.

McLeish (220) believes that the chromosome aberrations induced by x-rays may be influenced by the presence in the cell of additional nucleolar organisers. He finds that chromosome breakage seen in *Vicia* root tip cells 6-10 hours after x-irradiation is significantly less in those which contain three or more nucleolar organisers resulting from treatment 62 to 66 hours previously with maleic hydrazide. The cells less damaged by the hydrazide treatment which contained less than three nucleolar organisers responded to x-radiation in a manner not significantly different from untreated roots.

6. The Influence of Environmental Factors on the Response of Cells to Irradiation

Most of the investigations to be reported next have been prompted by consideration of the hypothesis that some part of the biological effects of radiation stem from the radicals formed in the aqueous phase. By no means all the observations are most easily interpreted in terms of radiation chemistry.

(a) *Water content.*—

Papers by Caldecott (49, 50, 51, 52), Konzak (191) and Ehrenberg (88), who worked with barley, all show that the influence of water content on the radiosensitivity of seeds may be much more complex than appeared hitherto, and does not necessarily point to the participation of radicals in the initiation of the biological damage. Caldecott & Konzak in general use seedling height at seven days as a measure of radiation damage to the seed, though this criterion is sometimes supported by observations of chromosome aberrations in the first root tip mitosis. In the first place Caldecott (50) confirms the previous finding that the sensitivity of seeds to x-rays is increased when their water content is increased by soaking at room temperature. It is also increased when the water content is increased by soaking the seed at 3°C., though maximum sensitivity in this case is not attained so quickly. When dry seeds are soaked at 0°C., however, sensitivity at first decreases with increasing water content, minimum sensitivity is reached after four to eight hours soaking, and by 24 hours the sensitivity is about back to that of the original dry seeds. The decreasing sensitivity with increasing water content is regularly seen when the water content is changed by equilibrating seeds with humid air.

In a second paper by Caldecott (51), a number of further points were investigated, and in particular the water content of embryo and endosperm were separately measured as a function (*a*) of the humidity of the air in which the seed was stored, (*b*) after soaking, (*c*) and after soaking and desiccation at different temperatures. From these additional observations it is concluded that sensitivity to x-rays, as measured either by inhibition of seedling growth or by chromosome aberrations seen in the first root tip division, is at a maximum when embryos contain about 4 per cent water and approaches a minimum when the embryos contain about 8 per cent water. Further uptake of water above 8 per cent to a level approaching 60 per cent does not increase x-ray sensitivity providing that water uptake occurs at a low level of metabolic activity. Growth inhibition resulting from exposure to a given number of fast neutrons per square cm. steadily increases as the water content of the seed is increased by soaking, but since the neutron dose in rads is mainly determined by the hydrogen content of the seed, it may well be that the damage produced by a given absorbed neutron dose rad is not much influenced by water content. The inhibition of seedling growth produced by a given number of thermal neutrons per square cm. was found to decrease with increasing water content, but again it may well be that this was for purely physical reasons, since the thermal neutron dose is in this case mainly determined by the reactions $^{14}N(np)^{14}C$ and $^{11}B(n\alpha)^{7}Li$.

Konzak (191) presents observations concordant with those of Caldecott in showing a large increase in the sensitivity of seeds to x-radiation as a result of soaking for 24 hours at 22°C. which is interpreted as due to physiological changes since the influence of soaking is smaller at low temperatures.

He finds that soaking at room temperature has very little influence on sensitivity to thermal neutrons. Konzak points out that even under conditions of controlled humidity, one embryo probably differs from another very considerably in water content, so that the difference between the influence of water content on sensitivity to x-rays and thermal neutrons may well account for the much greater variability in response of individual seeds to x-rays, compared with that to thermal neutrons.

Four criteria of radiation damage to barley seeds have been carefully studied by Ehrenberg (88), namely lethality, sterility, production of chlorophyll mutants, and chromosome abnormalities in the first root tip division, in an investigation of the influence of moisture content on damage induced by x-rays and fast neutrons. Starting with seed having 12.5 per cent water, the water content was either reduced or increased by controlled humidity for nine days at 23°C. The effectiveness of fast neutrons as judged by all criteria was found to be independent of seed moisture. Lethality, sterility and chromosome disturbances produced by x-rays were all found to decrease with increasing water content over the range investigated. Mutation rate decreased significantly between 9 and 11.5 per cent water content (0–30 per cent humidity) but not significantly between 11.5 and 18 per cent (30–100 per cent humidity).

Considering together the work of Ehrenberg, Caldecott & Konzak, it seems that a somewhat tangled situation is being clarified. In the case of slow and fast neutron effects on seeds, the amount of damage is essentially the same per rad, i.e., per unit of absorbed energy, in dry seeds and seeds which have taken up substantial amounts of water x-ray effects on the other hand are greatly influenced. The moisture content of the seed at the time of irradiation also influences response to post-irradiation treatments.

In the case of seeds, therefore, the water content itself appears, at least within certain limits, to be more important in setting the pace of physiological development than in supplying radicals.

It is interesting to compare these results with seeds and studies of hydration on an entirely different level. Rugh & Clugston (303) have taken advantage of the fact that in the ordinary course of nature eggs of the brine shrimp Artemia may dry up at the blastular stage but will develop normally when rehydrated, to study the influence of dehydration on radiosensitivity. Artemia was found to be considerably less sensitive dry than when rehydrated. Sensitivity was increased most by hydration for a period of one hour, possibly because more prolonged hydration resulted in the embryo developing into a more radioresistant phase.

(b) *Oxygen.*—

The work of the past decade leaves no room for doubt that oxygen may play a very important role in determining the outcome of exposure to ionizing radiation. The influence of oxygen on many different types of biological damage has been observed to conform to a pattern which has

much in common with its influence on the radiation chemistry of aqueous solutions.

A number of investigations published during the year tend to strengthen this association. Conger (63 and 64) has published a fuller account of the influence of oxygen on chromosome structural damage induced in mouse Ehrlich ascites tumour cells irradiated *in vitro* by x-rays and by neutrons than that which appeared formerly [Gray *et al.* (127)]. The relation found with x-rays is similar to that observed previously for chromosome structural damage induced in plants and insects. Neutron induced changes are little influenced by oxygen, with the result that the biological effectiveness of neutrons relative to x-rays itself depends on oxygen tension, varying from about 2.5 under aerobic conditions to 6 under anaerobic conditions.

It might be expected from a radiochemical standpoint that biological damage due to radicals formed by decomposition of water would be influenced by dissolved hydrogen as well as by dissolved oxygen, but such an effect has so far only been observed in connection with the indirect inactivation of bacteriophage. Read (286) in particular examined this point using growth inhibition in *Vicia* roots as a criterion of biological damage and worked with hydrogen at pressures up to 14 atm. but with negative results. Ebert & Howard (86) also using *Vicia* seedlings, have now observed that hydrogen at 50 atm. pressure reduces x-ray damage to roots both in the presence and absence of oxygen. Mortality was less when roots were exposed in 50 atm. pure hydrogen than in the same pressure of pure nitrogen, and x-ray inhibition of root growth was less in the presence of 50 atm. hydrogen added to 1 atm. air than under any of the following conditions.

The possible role of hydrogen peroxide production in radiobiology is briefly reviewed by Ebert (85).

Sufficient work has, however, now been done for complexities to begin to appear, both at the chemical and biological level. Exceptions have been found to most generalisations which have been made. In the first place Wolff & Luippold (363) appear to have established one situation in which anoxia immediately after irradiation can modify the nature of the chromosome aberrations to which the irradiation gives rise. In this case it was fairly clear that the determining factor was active respiration rather than high oxygen tension—indeed the simple displacement of oxygen by nitrogen or by evacuation was ineffective. The experiments consisted in exposing the soaked *Vicia* seeds to two irradiations, separated by a 75 min. interval, the first being a dose of 600r given in an atmosphere of nitrogen. When respiratory activity was reduced by chilling to 0°C., or stopped during the 75 min. interval, either by removal of oxygen and treatment with $Na_2S_2O_4$, by 2.10^{-3}M KCN or by 95 per cent CO plus 5 per cent O_2 in the dark, fractionated treatment gave many more aberrations in the first root tip division, three days later, than when seeds were normally aerated in the interval or were exposed to 95 per cent CO plus 5 per cent O_2 in the light. Treatment

with 1.5 or 3.10^{-4}M DNP in the presence of oxygen gave essentially the same results as treatment with respiratory poisons. Following the orthodox interpretations of chromosome exchange formation it was assumed that the larger number of aberrations resulted from unions between ends broken in the first and second irradiation, and thus indicated that healing had not taken place in the interval. From this it followed also that during the immediately post-irradiation period oxygen played a metabolic role in supplying energy through ATP for the fusion of broken chromosome ends.

Wolff & Atwood (362) had previously presented reasons for believing that the rate of healing of chromosome ends was controlled by an unidentified system subject to radiation injury independently of the chromosomes themselves and not simply by the degree of damage sustained by the broken ends. The state of the chromosomes studies by Wolff and Luippold may be more closely related to those of soaked barley seeds than to those of *Vicia* seedlings, upon which most previous work with *Vicia* has been done. Indeed Kihlman (174), who presents strong evidence for a metabolic role of oxygen in the induction of chromosome damage in *Vicia* root tip cells by 8-EOC, finds no indication of a similar effect with x-rays. Reference has been made elsewhere to Kihlman's studies of the chromosome structural damage induced in allium by x-rays and the radiomimetic chemical 8-EOC. This chemical agent is of special radiobiological interest not only because of the close quantitative resemblance between the aberrations which it induces and those induced by ionizing radiations, and the fact that like x-rays it is most effective when given in late interphase after DNA synthesis is believed to be complete, but also because its effectiveness is critically dependent on the presence of oxygen at the time of irradiation. This sharply distinguishes it from most other radiomimetics, including, for example, the nitrogen mustards. However, its mode of action cannot be identified with that of x-rays for several reasons. The radiomimetic effect of 8-EOC on *Vicia* meristem cells unlike that of x-rays is influenced by pre-treatment exposures to anaerobic conditions. It is not uniquely determined by oxygen tension but by the functional activity of the respiratory enzyme chain. Hydrogen peroxide supplied under anaerobic conditions enables 8-EOC to induce chromosome aberrations with an efficiency approaching aerobic effectiveness. It is believed that the importance of respiration consists in maintaining a supply of ATP essential to the radiomimetic action of this chemical, and this suggestion is supported by the fact that 8-EOC is inactive in the presence of dinitrophenol, which uncouples respiration and phosphylation, though it is active in the presence of ortho-nitrophenol. There remains, however, the fact that if from the total aberrations are subtracted those produced under anoxic conditions so as to obtain the number of aberrations induced by mechanisms involving oxygen, the curve for the influence of oxygen tension on aberration frequency is practically indistinguishable in form for 8-EOC and radiation induced aberrations.

Swanson (333, 334 and 336) has reported observations of different types

of chromatid aberration in *Tradescantia* which show the influence of oxygen at the time of irradiation to be different for four radiation qualities, viz. 50 kVp, 100 kVp and 250 kVp x-rays, and ^{60}Co gamma rays (1.2 mev). The oxygen/nitrogen aberration frequency ratios take the values 0.5, 0.83, 1.2 and 1.9 for chromatid deletions; and 1.9, 2.1, 2.2 and 2.3 for chromatid exchanges. The variation of aberration frequency with x-ray quality in itself poses a problem, quite apart from the superimposed influence of oxygen, which could be looked at in the light of a more detailed consideration of LET spectra (cf. Section 7) and the possible role of oxygen in extending the effect of length of an electron "tail" [Scott (311)] but it is not evident that order could be restored in this way. Swanson concludes that oxygen mainly influences rejoining of broken chromosome ends rather than primary break production. Conger (65), however, reports that in the same material (*T. microspore*) in which rejoinability of broken ends may be estimated from the incompleteness of isochromatid production and from the incompleteness of exchanges, no influence of oxygen is found in the former case, and a small influence in the latter. He concludes that it is quite impossible to account for the influence of oxygen on the observed aberration frequencies without allowing that oxygen influences primary breakage. Baker (17) and Baker & Von Halle (19) believe that in the case of *Drosophila* chromosomes oxygen operates almost entirely through an influence on the rejoinability of chromosome ends broken by the irradiation of sperm. They work with a stock carrying a ring X Chromosome and their experimental observations are that oxygen at the time of irradiation has little influence on the disturbance in the sex ratio or on the number of X-O males in the F_1 generation from crosses between untreated females and sperm from flies irradiated 0-24 or 24–48 hours before mating. Following orthodox interpretations of the consequences of breakage and reunion in a ring chromosome they conclude that if breakage frequency had been substantially influenced by the presence or absence of oxygen it would certainly have been detected. Baker & Edington (18) had previously observed a large effect of O_2 on the production of translocations between the chromosomes of *Drosophila virilis* and a moderate effect on induction of sex-linked recessive lethals in *Drosophila melanogaster* when the sperm were irradiated. Baker & Von Halle (20), however, found only a very small effect of O_2 at the time of irradiation of the sperm on the yield of dominant lethals in *D. melanogaster*. The mating procedures appear to have been comparable in these three experiments and in those of Baker & Von Halle with the ring chromosome of *D. melanogaster* referred to above. It may be noted, however, that in the two experiments which showed only a very small influence of O_2, the interval from irradiation to insemination never exceeded 2 days, and irradiation to fertilisation never exceeded 4 days since Sobels (322) has observed that administration of cyanide or azide prior to irradiation reduces the frequency of sex-linked recessive lethals in *D. melanogaster* very markedly in the four to seven day broods but barely significantly in the one to four day broods. A good im-

pression of some of the complexities of the subject will be gained by reading in sequence the papers presented by Giles (112), Swanson (334) and Baker & Von Halle (19), together with the contribution to the discussion by Conger (65) at the Oak Ridge Symposium on Genetic Recombination. The subject is also reviewed by Baker (17).

Advances in our understanding of the influence of oxygen are bound up with our knowledge of the mechanics of chromosome aberration production and of the oxygen tension which prevails in the immediate environment of the different cells composing an irradiated tissue.

Beam (24) brings forward evidence distinctly favouring the participation of oxygen in the initiation of genetic damage, rather than in the cell's ability to survive a given degree of damage. Using haploid, diploid and tetraploid yeast the influence of anoxia could be judged by six criteria, viz. survival of nonbudding cells and of budding cells in each of the three ploidys. By all six criteria cells irradiated in air were just about twice as sensitive as cells which were equilibrated with nitrogen for the period of irradiation (extreme values of the air/nitrogen ratio were 1.8 to 2.4). This constancy is the more remarkable in view of the probable difference in mechanisms responsible for the failure of the cells of different ploidys and of different phase within each ploidy to survive and form colonies.

Mention has already been made of the fact that oxygen does not increase the inactivation of pneumococcal transforming principle (now generally accepted to be pure DNA) when irradiated in aqueous solution under conditions which expose it to attack by radicals. The virus S.13 (largely if not entirely DNA) may be either more or less inactivated in the presence of O_2 according to the interval between irradiation and assay. Alper (5) gives a much fuller description than has appeared previously of her studies of the inactivation of dilute suspensions of the bacteriophage S13 by gamma, electron and x-radiation. The experiments are specially concerned with the influence of dissolved gases, including oxygen, hydrogen and mixtures of these gases, and are analysed from the standpoint of radiation chemistry. Though survival is always assayed by the ability of the phage to form plaques when plated with a sensitive organism, three forms of injury can be distinguished: 1, complete inactivation during irradiation; 2, part inactivation during irradiation which is revealed by susceptibility to subsequent inactivation by various agents, including H_2O_2; 3, delayed inactivation during the post-irradiation period through interaction between part inactivated virus and H_2O_2 formed during irradiation. When inactivation is assayed immediately after the end of irradiation the greatest yield is obtained when the suspension is bubbled with a mixture of hydrogen and nitrogen, least when bubbled with hydrogen and oxygen, and intermediate when bubbled with oxygen and nitrogen—these being just the reverse order of effectiveness to that for H_2O_2 production by irradiation of water. It is concluded from these and other observations that phage is not inactivated by OH or HO_2, but by H and in certain circumstances by O_2^-. Part inactiva-

tion is also believed to be due to reducing radicals, and varies in importance with the dose; the number of virus particles partially inactivated falls from 10 times the number fully inactivated at 1000 rad to 1/30th at 20,000 rad. Thus, for the inactivation of this virus, oxygen present at the time of irradiation, increases the overall damage only through the formation of H_2O_2, which subsequently completes the inactivation of injured virus particles.

Fresh experimental evidence has been brought forward both for and against the view [Gerschman *et al.* (108)] that there exists a real analogy between the mode of action of ionizing radiations and oxygen poisoning in mammals. Plaine (267) has conducted experiments which he interprets as strengthening the biological effects of ionizing radiation and of chemical treatment with H_2O_2 and exposure to elevated concentrations of oxygen. Following previous work [Glass & Plaine (116), and Plaine & Glass (269)] two biological reactions are studied, namely the expression of an eye abnormality, "erupt eye" in the strain in which this abnormality is normally absent on account of a suppressor gene, and the development in the larvae of what the authors consider to be melanotic tumours. Both effects are produced by exposure of the ten hour *Drosophila* embryo to x-rays, and the x-ray damage shows the usual dependence on oxygen tension. Only melanotic tumours had previously been produced by elevated oxygen tension alone (10 minutes). Subsequent work has shown that one hour exposures in an atmosphere of O_2 produce the erupt manifestation, and that when two strains of *Drosophila* are compared, that which is the more sensitive to x-rays is also more sensitive to oxygen. Positive effects, somewhat obscured by general toxicity, have also been obtained by treatment with H_2O_2. Mention is made of the fact, supported by experimental results presented in a subsequent paper (268) that both manifestations of damage induced by x-rays are greatly reduced by cysteine, not only when present at the time of irradiation, but when fed to the larvae after irradiation has been completed.

In conformity with observations on many other materials, Clark & Herr (57) have found that the sensitivity of Habrabracon pupae at different stages of development and of larvae in cocoons was reduced by a factor of about 3 when irradiated in an atmosphere of nitrogen instead of air. Larvae irradiated in hydrogen, however, were somewhat more sensitive than those irradiated in nitrogen. It is pointed out, however, that the analogy between oxygen poisoning and radiation damage in mice [Gerschman, Gilbert, Nye Dwyer & Fenn (108)] breaks down for Habrabracon since pupae which are very sensitive to oxygen poisoning and larvae which are insensitive show the same influence of oxygen on x-ray sensitivity.

Using death of Paramecium after autogomy as a measure of genetic damage Kimball, Hearon & Gaither (178) have compared the effect of irradiation and of immersion in hydrogen peroxide. The question of the access of externally applied hydrogen peroxide to the micronucleus, considered as an open system, i.e., without reference to possible membrane

impermeability, was evaluated theoretically [Hearon & Kimball (137)] from the geometrical form of the organism and the measured rate at which it destroys hydrogen peroxide. Since externally applied H_2O_2 at concentrations up to 0.1M/litre, and short exposures to still higher concentrations, were without effect, it is concluded that the mutagenic effect of x-rays is not mediated through H_2O_2. Moreover, H_2O_2 treatment after irradiation in nitrogen was also without mutagenic effect [as reviewed by Kimball (177)].

The rational for increasing partial pressure of oxygen in respired gas in order to improve the sensitivity of tumours differentially as compared with normal tissue [Gray, and co-workers (127)] rests on the ascertain relation between radiosensitivity and oxygen tension for many different types of cell, including tumour cells, and on the supposition that some parts of tumours are more anoxic than most normal tissues. Thomlinson & Gray (339) find quantitative support for this supposition in the histological structure of many specimens of human carcinoma of the bronchus. Tumour cords, lacking any capillary structure, derive their nutrients, including oxygen, from the surrounding stroma. Approximate calculation of the oxygen gradients through such cords shows that necrosis sets in at just about the distance from the stroma at which oxygen tension would be expected to have fallen to zero. This distance, which is approximately the same for all cords irrespective of their size, is 100 to 150μ. It is felt that insufficient allowance has often been made for such oxygen concentration gradients in interpreting radiobiological response to changes in environmental oxygen tension. For example enzyme poisons, by depressing or stopping cellular respiration, will reduce any gradients which exist and elevate O_2 tension at a distance from the source [cf. Hall (134a)]. This mechanism has therefore always to be considered when an enzyme poison is found to increase radiosensitivity. It was one of the possibilities considered by Sobels (322) to account for the effect of cyanide and azide injected into the abdomen 1–30 mins. before irradiation in increasing the sex-linked recessive lethal mutations induced in *Drosophila* sperm by x-rays.

In connection with the clinical use of oxygen to increase the radiosensitivity of tumours, Howard-Flanders & Wright (158) have examined the reduction in growth rate of the tails of very young mice brought about by exposing the tails to 1500 to 3000r of x-rays, either when the blood supply to the tail was clamped off during irradiation, or when the partial pressure of oxygen in the gas respired by the animal was 0.1, 0.2, 0.4, 1, or 3 atm. At 1 atm. of oxygen, radiosensitivity was estimated to be 1.30 ± 0.06 relative to that when the mouse was breathing air. Though less than the difference found for tumours, this factor is a considerable one with obvious clinical implications. It is of interest that the layer of dividing cells which control the bone growth is separated from the nearest blood vessels by some 100μ of metabolising tissue which would probably establish an oxygen gradient similar to that believed to exist in some tumour cords. The situation

is therefore one in which an increase of radiosensitivity with partial pressure of oxygen in the respired gas is to be expected.

Limiting oxygen gradients may even arise in concentrated actively matabolizing cell suspensions. Goucher, Kamei & Kocholaty (121) find that protection of *E.Coli* against x-radiation by pre-incubation with substrates such as succinic acid and ethanol occurs only with dense bacterial suspensions, in which respiration appears to limit equilibration with the surrounding oxygen. Protection is found to be abolished altogether by shaking the suspension for a few seconds before irradiation. A given dose of radiation resulted in essentially the same per cent, inactivation for suspensions ranging from 10^4–10^9 cells per ml. provided that oxygen was freely available.

The influence of cold, HCN and anoxia at the time of exposure of fertilised uncleaved salamander eggs to x-rays has been tested. With respect to embryo abnormalities and viability, cold gave significant protection and anoxia very significant protection, but HCN was without effect [Peters (263)].

The effects of respiratory enzyme poisons administered before or after irradiation of grasshopper eggs containing embryos in diapause have been studied by Tahmisian & Devine (338) and Tahmisian (337). The area of the embryo 16 days after irradiation was used as a criterion of radiobiological damage. The results of treatment with respiratory poisons were complex, but appeared to include some protective action resulting from post-irradiation treatments with cyanide.

Altenburg (6) has observed a striking synergism between ultraviolet and tertiary butyl hydroperoxide in producing lethal mutations in the second pair of autosomes of *D. melanogaster*. Since shielding of the polar cap regions from ultraviolet reduced mutation frequency to that of the tertiary butyl hydroperoxide alone, the syngergism is presumed to be intracellular.

Giese *et al.* (111) have reviewed 100 papers bearing on the question as to whether the effects of ultraviolet in killing bacteria, retarding cell division, inducing mutations and activating eggs, are mediated through the production of peroxides, and attempt to resolve the question by looking for analogies and synergisms between the effects of ultraviolet and H_2O_2 on protozoa, on the gametes of the sea urchin, and in inducing changes in the absorption spectra of some of the major constituents of cells. They have also attempted to influence ultraviolet damage by catalase and glutathione. Differences between the actions of externally applied H_2O_2 and exposure to ultraviolet were more prominent among their results than similarities, but the question was considered still to be an open one.

It cannot be said that the year's work has settled any of the major issues. Cases in which the role of oxygen is at least in part metabolic are making their appearance. In view of the importance of oxidative processes as a source of energy and the likely need for energy in the development of

radiobiological lesions it is remarkable that the metabolic aspect is still not very prominent.

Laser (203) has examined the influence of oxygen on the sensitivity of a variety of bacteria, including strict as well as facultative anaerobes. Most of the bacteria examined were more sensitive when irradiated in the presence, than in the absence, of oxygen, but *Sarcina lutea* presented certain unusual features. The influence of oxygen on the sensitivity of this organism could be suppressed completely by a variety of enzyme poisons, CO, KCN, hydroxylamine, and azide, known to combine with oxidised cytochrome a^3, but was not affected by urethane. It was concluded that at least for this organism, the enhancement of radiation damage involves the respiratory mechanism and requires that part of the respiratory chain should be in the oxidised state.

In many other organisms and tissues there is no indication at all so far as to what part, if any, the respiratory system plays in the initiation and immediate development of damage. Instances have been cited elsewhere [Bellamy & Lawton (26, p. 12)] of strains of bacteria deficient in respiratory enzymes, which show a 3:1 increase in sensitivity when irradiated in the presence of oxygen, and of a strain of yeast ("petite," under (*a*) *Action spectra*) deficient in respiratory enzymes, which has the same sensitivity as the normal strain.

(c) *Chemical pre-treatment.*—

The fact that the composition of a tissue may greatly influence the absorbed dose in rads delivered to the cells of that issue when it is exposed to x-radiation has been fully appreciated for some time and experimentally demonstrated [c.f. Sigmund & Witte (316) in the current literature]. Much larger differences probably arise in tissues exposed to thermal neutrons, but these are harder to assess because they depend critically on the distribution of nitrogen, boron and possibly lithium, within the irradiated cell itself. It has not been found practicable to influence significantly the absorbed x-ray dose to a tissue by chemical treatment, but it is now 16 years since Kruger (194) first demonstrated that the high cross section of boron for slow neutrons could significantly increase the energy absorption in tissue in which the boron was incorporated as borax. Boron, especially B^{10}, significantly increases chromosome structural damage in microspores exposed to slow neutrons, and the therapeutic possibilities of boron sensitised slow neutron irradiation are under clinical investigation. It has now been shown [Kruger (195)] that three boron containing dyes administered to mice intravenously show a 30 fold greater concentration of boron in the growing edge of an anaplastic mouse brain tumour than borax.

In cases where the sensitivity of cells to x- or γ-radiation is influenced by a chemical treatment some reason other than a change in absorbed dose must be sought.

(i) Investigations in which treatment is reported to decrease the effectiveness of an irradiation: it is now a well established fact that pre-treat-

ment of micro-organisms, single cell suspensions, plants, tissues and whole animals, by the cysteine, cysteamine, and cystamine group of substances and by thiourea may under certain circumstances diminish the effectiveness of an exposure to x- or γ-radiation. Protection has also been reported against cellular damage in animals to which these chemicals have been administered. We may therefore note the observations of Langendorff, Koch & Hagen (202) that after injection of 30 mgm. thiourea per 20 gram mouse the lungs become inflamed to an extent likely to give rise to hypoxaemia. Bacq, Cuypers, Evrard & Soetens (16a) have observed a drop in the average percentage saturation of venous blood of rats following administration of cystamine. Such a hypoxaemia might clearly be an important element in all protective effects of such administrations. Leaving aside the physiological effects of the administration of these compounds to animals which are more especially of concern in connection with the response of the animal to whole body irradiation, there are nevertheless several chemical and metabolic aspects which have to be considered: (*a*) cysteine and cysteamine, more especially on the alkaline side of neutrality, readily combine with molecular oxygen, and measurements of the rate of removal of dissolved oxygen from solutions under some of the conditions used in radiobiological studies with bacteria, cell suspensions and roots [cf. Gray (126)] make it probable that a degree of hypoxia has often been induced which is adequate to account for the observed protection. This could more frequently be controlled than has been the case hitherto. It is noteworthy that these chemicals have not in general been reported to afford better protection of isolated systems than anoxia. An exception must be made in the case of the protection of bacteria by cysteamine and its derivatives, where the efficacy of pre-treatment is linked with the nature of the post treatment in such a way as to suggest that these compounds may be of importance to the development as well as the initiation of the injury (153). (*b*), cysteine is a natural amino acid, but is deleterious, and even lethal to some organisms when present in food at 10mM concentrations [Plaine (268)]. Cysteamine, but not cystamine or cysteine, according to Koch (187) accelerates the growth of Ehrlich ascites tumours; (*c*), cysteamine undergoes spontaneous oxidation in the presence of serum proteins and cystamine reacts with proteins to give cysteamine. We owe much of our knowledge of this important aspect of the chemistry of cysteamine and cystamine, as well as of the radiation chemistry of these compounds to Eldjarn, Pihl & Shapiro who have summarised the present position (91).

Some new features of the protection of *E. Coli* B/r by cysteamine and β-mercaptoethanol have been revealed by the experiments of Hollaender & Doudney (153). β-mercaptoethanol given before irradiation, though slightly the less effective of the two for doses up to 60kr, affords the simpler type of protection in that the dose reduction factor is about 8 for all doses up to 120kr and is not much affected by the medium on which the bacteria are cultured after irradiation. The effectiveness of pre-treatment with cysteamine is critically dependent on the post-irradiation treatment. When cells

which were previously cultured in a rich medium are given optimum survival conditions after irradiation the dose reduction factor for pre-treatment with cysteamine is 12, but this falls to 2 if cells are plated on minimal agar after irradiation. However, cells which have been in contact with yeast extract for only 15 minutes after irradiation, before plating on minimal agar, show full cysteamine protection. The cysteamine appears to act intracellularly, since cells which have been pre-treated for 30 minutes remain highly resistant even if washed before they are irradiated. There is no close correspondence between the efficiency of a variety of compounds tested with respect to protection against bacteria and mice; in particular S-β-aminoethylisothiuronium-Br-HBr, which Doherty & Burnett (82) find to give better protection to mice against whole body X-irradiation than any of the other -SH compounds, is completely ineffective for bacteria. It is also found that the frequency of induction of reverse mutations in an adenineless strain of *E.Coli* is depressed by a given treatment with cysteamine or β-mercaptoethanol to the same extent as survival is enhanced, a relation previously found to be true for anoxia with this particular mutant. The influence of temperature of incubation on the induction of these mutations has been studied by Anderson & Billen (8). There seems to be no influence of an added suspension of radiation sterilised cells on mutagenesis of unirradiated cells. Mikaelsen (230, 231) (Section 5) has studied the influence of protective chemicals against low dose rate irradiation. *Traxescantia* roots were exposed in nutrient solution a total dose of 50r of Co^{60} γ-rays delivered in 48 hours. Roots were examined at the end of irradiation. The proportion of abnormal anaphases was reduced when glutathione, cysteine, thiourea or $Na_2S_2O_4$ were added to the culture medium. Of the compounds tested cysteine was the most effective, giving better protection when added to the culture medium as the hydrochloride at 0.5mM concentration than when neutralised. This may be related to the rate of autoxidation of this compound, which might be sufficiently rapid in alkaline solution to eliminate the reduced form of the compound early in the exposure. Good protection was observed with thiourea, which at 10^{-3}M reduced the proportion of abnormal anaphase figures seen at the end of exposure by 48 per cent. This appears to be the largest biological protective effect so far reported for thiourea, and it is of interest that it has been obtained in conjunction with very low dose rate exposure, since Dale (70) has recently shown that when sufficiently low dose rates are used the ionic yield for the decomposition of thiourea in aqueous solution rises to values of over 10,000 as a result of chain reactions. These chain reactions only occur in the presence of oxygen, and oxygen is consumed in proportion to the thiourea decomposed. An induced intra-cellular anoxia should therefore perhaps be added to the other possible explanations of the protective effect which was observed. The frequencies of chromosome interchanges and deletions in onion root tip cells resulting from a given dose of x-radiation has been found by Riley (293) to be reduced when the roots are immersed in 2.10^{-3}M sodium hydro-

sulphite or 2.10^{-4}M BAL for 30 minutes before, during, and for 10 minutes after irradiation. Protection was just as great when the roots were in either solution before and after, but not during, irradiation. Protection of *Vicia* seedling roots against x-ray induced chromosome structural damage by addition of a colloidal preparation "Kollidon" to the culture medium is reported by Burger & Peters (61). Protection just reaches the conventional level of statistical significance when the average number of abnormal anaphase figures in treated and untreated roots are compared 12 to 30 hours after irradiation. Devik & Lothe (79) have compared the effects of anoxia and chemical protection with respect to early signs of cytological damage in bone marrow, of mice which have received whole body irradiation. Some mice survive 1100r whole body x-irradiation if pre-treated with cysteine, cysteamine and cystamine, but survival is much less than when animals are breathing 8.5 per cent oxygen at the time of irradiation, and pre-treatment with the chemical agents scarcely adds to the protective effect of hypoxia. Cytological damage to bone marrow was less in all combinations of protective procedures, tested but the variability was such that differences were either not significant or on the borderline of significance. Moreover, humoral effects cannot be ruled out in such cases (cf. Section 4a).

In a second investigation Lothe & Devik (217) studied the influence of cysteamine and anoxia, alone and in combination, on the response of the skin of the rabbit's ear to localised X-irradiation; three experimental procedures were adopted; (*a*) a ligatured ear of a rabbit was perfused with nitrogenated Ringer solution and the other used as control (anoxia); (*b*) one ear was perfused with the same solution containing cysteamine at 3 or 100 mgm. per cent, the other being perfused with the nitrogenated Ringer solution (anoxia plus cysteamine); (*c*) one ear was perfused with citrated oxygenated human blood containing 100 mgm. per cent cysteamine, and the other with the oxygenated blood only (cysteamine). The local skin reactions were in all cases reduced by anoxia. Cysteamine alone also reduced skin reactions, though early reactions were more affected than late reactions. Cysteamine doubtfully added to the protective effect of anoxia. An aqueous solution of cysteine injected subcutaneously has been observed to afford protection against x-ray induced erythyma reaction in the rat tail [Hofmann (144)]. Intraperitoneal injections of 15 mgm. cysteamine into 200 gram rats one minute before local exposure of the anterior upper abdomen to 500r of x-rays has been observed by Desaive & Varetto-Denoël (75) to decrease radiation induced mitotic inhibition in the intestine. An extensive comparative study of cytological damage to the intestine by x-rays and by nitrogen mustard by these authors included observations showing that cysteamine given three minutes or ten minutes before nitrogen mustard diminishes the extent of cytological damage, but it does not do so if given simultaneously or immediately after the nitrogen mustard. Neither cysteamine at 4 μg/gm. body weight, which is the maximum tolerated by the silkworm moth when injected into the abdomen, nor 40 μg/gm. of cysteine, was observed to

protect against either x-ray induced mutation affecting egg colour or against hatchability [Nakao, Tazima & Sugimura (245)].

The mode of action of protective chemicals, more particularly those of the -SH group, has been briefly reviewed by Patt (258), who has presented and discussed elsewhere (259) some peculiar features of the influence of oxygen, cystein, pH, and temperature on the survival of irradiated lymphocytes. The literature on the protective effect of cysteine against many forms of radiation damage has also been reviewed by Hofmann, and co-workers (145). In Oehlert's experiments with barley seedlings and Urodel larval cornea (253), cysteine is observed to protect against x-rays, but no information is given as to the influence of the cysteine on the oxygen tension prevailing at the time of irradiation, nor are the experimental details given in the paper sufficient to form an opinion on this point.

(ii) Investigations in which an enhancement of radiobiological damage has been reported: several cases of enhancement of radiation damage by chemical pre-treatment have been reported. Figge & Wichterman (98) have reported an extremely marked enhancement of radiation damage to *Paramecium caudatum* when animals are exposed to x-rays in the presence of as little as 1:100,000 haematoporphyrin. Since the shape of the survival curves was altered, no dose amplification factor can be given, but the published curves show 50 per cent survival after 340,000r in untreated animals and about 18,000r in treated animals. The animals were irradiated in plastic windows and were probably anoxic at the time of irradiation. A remarkable feature was that the animals irradiated in the presence of porphyrin to dose levels of 20–80,000r only died when brought into contact with oxygen.

Haematoporphyrin is one of many substances which sensitize these organisms to killing by visible light (photodynamic action). Steps were taken to exclude such action by keeping the organisms at an appropriately low intensity of illumination. It may be noted, however, that it is characteristic of some photosensitised oxidations [e.g., riboflavin-sensitized oxidation of auxin, Galston (104)] that they may take place on admitting oxygen at some time after illumination.

10^{-3}M Malonic acid, which alone does not depress colony formation from single cells of Saccharomyces ellipsoideus, has been observed by Kigs, Ando & Koike (173) to depress the survival after 10 to 50,000r of x-rays, if applied before irradiation. This enhancement of radiosensitivity was entirely offset by the addition of 2×10^{-3}M fumaric acid or 10^{-5}M aspartic acid to the Nägeli agar on which the cells were cultured for 20 hours after irradiation. Calcium deficiency, which appears to exert a quite definite influence on "spontaneous" chromosome aberrations, micronuclei formation and pollen abortion in Tradescantia paludosa, is reported by Steffenson (330) also to increase sensitivity to x-rays. Post-irradiation treatment with Mg. ions has previously been observed [Daniel & Park (71)] to reduce radiation damage to Hydra.

Mitchell (233) reports a significantly increased proportion of tumour regressions in rats bearing Walker carcinoma 256 when x-radiation was combined with Synkavit administered intravenously. This corresponds with clinical experience of improved response in patients suffering from carcinoma of the bronchus. No beneficial results were obtained when the Synkavit was administered intramuscularly. In discussing this paper Neukomm (247) summarises observations by himself and his colleagues which may in part explain this difference between intravenous and intramuscular administration since he finds that Synkavit is readily adsorbed on to protein and cellular membranes. Kohn & Gunter (189) have observed no influence of Synkavit on the sensitivity of either *E. Coli* or yeast to x-rays. Peters, Gärtner & Krais (262) report a synergism between γ-radiation and Megaphen (N-(3′-dimethylamino)propyl-3-chlorphenothiazine) as regards cytological damage to Ehrlich ascites tumour cells. Since the whole animal was exposed to the action of both drug and radiation, the effects observed in the ascites tumour cells may have involved response of the whole animal. However, a synergism had previously been observed in *Vicia* roots.

In a brief note, Kaplan (166) reports large differences between the mutagenic and lethal effects of ultraviolet on *B. prodigiosum* according to the moisture content of the gas with which the cells were in equilibrium at the time of irradiation. Lethality and mutation rise steeply and in proportion by a factor of 20 as the humidity falls from 97 per cent to 33 per cent. Both effects are photoreactivable, but greater survivals are only observed if the humidity is high during both ultraviolet and visible light exposure, whereas the photoreactivation of mutation only requires that the humidity should be high during ultraviolet irradiation. This difference might be connected with the further observation that light alone inactivates dry, but not humid, cells.

(d) *Investigations concerned with post-irradiation treatments.*—

(i) Chemical and metabolic factors. Radiobiological damage is rarely, if ever, expressed unless there has been metabolic activity in the interval between irradiation and observation. In some cases the immediate post-irradiation period appears to be particularly critical. It may be that a very important aspect of post-irradiation restoration has been opened up for investigation by Ungar, Damgaard, & Williams who have observed that intraperitoneal injection of rat spleen homogenates 30 min. to 4 hrs. after exposure of the skin to Sprague-Dawley rats to 3000r delays the onset and reduces the severity of the resulting skin lesions. No systemic effects were observed at any time. Liver homogenates injected under the same conditions were not observed to bring about any restoration. The importance of a supply of energy-rich phosphate bonds during the interval between two fractionated exposures of soaked seed upon the yield of chromosome aberrations has already been considered (363). The experiments reported by Stepleton, Sbarra & Hollaender (329) are specially concerned with the nature of the metabolites which must be added to minimal media to secure maximum survival of

irradiated bacteria. It will be recalled that if *E. coli* B/r are cultivated on a rich medium before irradiation, but plated on to a minimal medium after irradiation and grown at 37°C, many fewer organisms survive than is the case if they are plated on to a rich medium and held for 24 hours at 18°C. before being grown at 37°C. A long search revealed that glutamic acid, guanine, and uracil, in concentrations of about 750 gamma/litre, 150 gamma/litre, and 150 gamma/litre respectively, added to agar containing glucose and salts gave the highest survival of all combinations tested. The type of medium used to culture the cells before irradiation, however, determines in large measure the composition of that which is best for the post-irradiation incubation, and it is suggested that the recovery process is related to the synthesis of new enzymes in the irradiated cell. The restoration of ultraviolet irradiated *E. coli* B/r has been studied by Heinmetz & Lehman (139). When bacteria were exposed to 2000 ergs/mm.^{2} ultraviolet radiation, incubated with metabolites of the tricarboxylic acid cycle and co-factors, extensive restoration of viability was observed. Metabolites or co-factors alone were less effective, and amino acids were not in general effective. Combined with photo restoration still higher survival values were observed. Pratt, Moos & Eden (279) have observed greater survival of irradiated *E. coli* after storage at 4°C. to 6°C. than in organisms plated immediately. This is most marked when organisms are diluted with broth after irradiation, but survival appears to be significantly greater than in controls plated immediately when storage is in citrate or even in distilled water, when allowance is made for the adverse effects of storage in distilled water at low temperature on untreated bacteria. That some recovery should be observed in the absence of nutrients probably does not conflict with the observations of Stapleton, Billen & Hollaender (328) since the recovery observed by Pratt *et al.*, represented by a difference between 10 per cent 40 per cent survival, would correspond to only a small dose reduction factor, and only became evident after storage for many days.

Bachofer & Pahl (16), following up the old observations of Cook (66), have studied the influence of post-irradiation temperatures on cleavage delay and embryogenesis in irradiated *Ascaris lumbricoides suum* eggs. Cleavage delay was found to remain unchanged when irradiated eggs, held at 0°C. and 5°C., for periods up to 35 weeks were subsequently incubated at 30°C. in order to initiate cell division. On the other hand, the proportion of embryos which developed normally after doses of 12,000r to 50,000r fell continuously with the duration of low temperature treatment. Temperatures above as well as below 30°C. were investigated. The pattern of survival is complex and probably influenced by the fact that low temperature alone without irradiation diminishes survival, but it is concluded that for maximum survival the post-irradiation temperature should aproximate that for maximum biological activity. Gaulden & Kokomoor (122) have observed an influence of culture conditions on the length of time during which irradiation delays the entry of cells into division. They find that the proportion of grasshopper

neuroblasts in a hanging drop culture which are in mid-mitotic phases at any time increases more than twofold with addition to the culture medium of up to one-half the normal yolk content of the egg, and that the re-entry of cells into this phase of mitosis after 64r of x-rays occurs much earlier in the presence than in the absence of yolk.

Reference has been made earlier to the effects of two suppressor genes in *D. melanogaster* whose action is inhibited by irradiation of the developing embryo in such a way as to give rise to flies with erupt eyes or larvae with what are taken to be melanotic tumours. These two forms of radiation damage are the more interesting because they are greatly reduced by cysteine given after, as well as before, irradiation—apparently the only case so far reported of an effective after-treatment with cysteine since Patt, Blackford & Straube (260) showed some protection of rabbit thymocytes by addition of cysteine immediately after an *in vitro* irradiation. The experimental procedure used by Plaine (268) consisted in adding L-cysteine to the yeast agar food at a concentration of 0.1 per cent for the whole of the pre-treatment period (24 hours in some experiments and 55 in others) or for the period between treatment and pupation, viz., about 75 hours and 44 hours respectively in the two experiments, or continuously before and after treatment for the entire period of about 100 hours. All three treatments reduce the effectiveness of the irradiation. Two other observations are also relevant: (*a*) the X-ray treatment alone and the cysteine treatment alone each result in considerable lethality (20 per cent and 25 per cent hatching respectively) but the situation does not defy analysis because deaths occur at different stages—with X-rays mainly between pupation and hatching, being due in many cases to gross morphological abnormalities, and with cysteine mainly before pupation. Continuous cysteine treatment, before and after irradiation treatment, very significantly reduced x-ray mortality between pupation and hatching, despite the toxicity of the cysteine at the earlier stage; (*b*) another amino acid, tryptophane, acting at a concentration of 0.5%, which is about half the 100 per cent lethal concentration, counteracts the effects of both suppressor genes and gives erupt eye flies and melanotic tumours in larvae. The effect of tryptophane is enhanced by high oxygen tension and is diminished to an extent comparable with the diminution of x-ray induced effects by addition of L-cysteine (0.1 per cent). The protective effect of cysteine against tryptophane was in all respects similar to that against x-radiation.

We have already commented (p. 40) upon several papers concerned with the effect of water uptake prior to irradiation upon the sensitivity of barley seed to x-rays and to fast and thermal neutrons. When the effects of post-irradiation treatments are studied [Ehrenberg (87)] further important points are brought to light. Damage induced by exposing dormant barley seeds to x-rays is increased when the seeds are stored by six weeks, more especially if they had a high water content when irradiated. Damage is also greater when the seed is sown at 12°C. than when sown at 25°C., and the effects of storage and temperature of germination are additive. When,

however, seeds which have already germinated are exposed to x-rays the temperature at which they are sown does not influence radiation damage; moreover they are not affected by storage if dried again after irradiation, prior to storage. It is evident that different mechanisms are operative in the development of radiation damage according to whether the seed is dormant or germinating at the time of irradiation. Damage induced by neutron irradiation is almost independent of all these post-irradiation treatments and must therefore differ in important respects from that induced by x-rays. Adams, Nilan & Gunthardt (1), also working with barley, confirm the earlier observations of Gustafsson (132) that x-ray damage is increased when seed is stored for up to eight weeks before germination. There were some indications that as regards chromosome damage in first root tip division, germination, emergence in the field, and seedling height at two weeks, radiation damage was enhanced more by storage in oxygen than by storage in nitrogen, but differences were only significant at the 5 per cent level in the case of one criterion, viz., emergence in the field. Bilquez (30) has found no effect of storage on the viability of irradiated chicory seeds (Crepis). Water content of the seed, temperature, germination and other factors, however, greatly influenced sensitivity to x-rays, as has been found to be the case with barley (31).

(ii) Photoreactivation. Photoreactivation remains a form of restoration peculiar to damage induced by ultraviolet radiation. Dulbecco writes authoritatively on this fascinating subject in Chapter 12 of Hollaender's *Radiation Biology* with literature references to the end of 1952. The action spectrum of the reactivating light shows that the absorbing molecule is generally a pigment, and could in the opinion of Dulbecco be the same pigment in the phage T-2, in *E. coli* B/r and in *Streptomyces griseus* spores, but the site of the damage which is reactivated appears almost always to be nucleoprotein. In conformity with this Von Borstel & Wolff (351) find that the lethal action on Habrabracon of a ultraviolet irradiation which includes the nucleus is photoreactivable while death due to irradiation of cytoplasm only is not. Kaplan's studies of the influence of moisture on the photoreactivability of *B. prodigiosum* have already been referred to (165).

Four other papers remain to be mentioned. Localised irradiation of regions of the forelimb of larval amblystoma gives rise to regression of the limb and induction of an accessory structure at the site of exposure. The extent of regression and the incidence of accessory structures are subject to photorecovery, both being markedly reduced by illumination by visible light for a few days after exposure to ultraviolet radiation [Butler & Blum (47)]. Photorecovery from ultraviolet injury has frequently been observed in lower forms and in plants; it was observed in vertebrates after repeated exposures, and in the developing forelimb of *Amblystoma* after a single exposure. Frog larvae exposed to single or repeated doses of ultraviolet and placed in total darkness developed pigmented streaks dorsally and isolated melanophores on the periphery of the cornea which appeared to be due to

local cellular lesions. Irradiated animals placed under visible light did not develop these lesions [Zimskind & Schisgoll (367)]. Photoreactivation, at least in its most general connotation of the effect of visible light in abating an injury due to ultraviolet radiation, appears to be reported for the first time in a mammal by Rieck & Carlson (291). The criteria used were survival of mice, and extent of damage to the ears, after whole body exposure for about 40 minutes daily to $1.6.10^8$ ergs/cm.^{2} of 200–313Mμ radiation. By both criteria damage was less marked in animals kept in strong visible light between exposures, compared with controls kept in the dark. On the other hand, Griffin and co-workers (128) report that if white mice are kept in the presence of light, the amount of ultraviolet energy per square cm. given as a daily exposure which is needed to elicit a given incidence of skin tumours, is slightly less than if the animals are kept in the dark, i.e., the light here appears to be operating in the opposite sense of a photorestoration.

7. The Influence of Linear Energy Transfer (LET) on the Biological Effectiveness of Ionizing Radiation

From the fact that the form of LET spectra derived from an analysis of the size of pulses of ionization in a chamber exposed to neutron radiation are not independent of the linear dimensions of the chamber, Rossi & Rosenzweig (301) conclude that LET spectra will similarly depend on the size of the biological structure irradiated. A still more serious limitation of the LET concept arises from the fact that biological effectiveness often increases sharply with LET in the range 6–30 kev/μ. Some of the energy of even the fastest electron is dissipated by energy transfers which give rise to δ-rays having an LET in this range. A statement that the LET of fast electrons is 0.2 ekv/μ, meaning that the average rate of loss of energy by the electron when all losses are considered is 0.2 ekv/μ, obscures the fact that a proportion of these losses, probably accounting in all for rather more than 10 per cent of the energy, give rise to highly effective slow electrons (δ-rays). The contribution of these small energy transfers to chemical and biological effects of radiation has been evaluated (123). When this is done, it is seen that the x-radiation from a tube operated at 250 kv may differ quite significantly in biological effectiveness according to whether it is filtered through a few millimetres of aluminium or by a Thoraeus filter. In the light of these estimates of biological effectiveness the differences in frequency of *Tradescantia* chromosome aberrations observed experimentally by Kirby-Smith & Daniels (183) with the two qualities of x-radiation referred to become intelligible [Gray (124)]. The same considerations might throw light on the relative effectiveness of x-rays generated at 140 kv, 200 kv, 1.25 Mv and 25 Mv x-rays with respect to chromosome damage in *Tradescantia* microspores which Arnason & Morrison observed to be roughly in the ratio 1.0:9.2:0.73:0.68. The number of electrons set in motion per unit volume and per unit dose by 200 kev, 1.25 Mev and 25 Mev x-rays were estimated to be in the ratio 1:0.024:0.006, which must also be the relative probabilities that a chromo-

some is traversed by an electron "tail" when exposed to equal doses of the three radiations. Since these figures bear no relation to the relative biological effectiveness of the three radiations, the authors conclude that Lea's hypothesis regarding the LET requirements for the induction of chromosome structural damage is invalid. The conclusion does not, however, follow if account is taken of the small energy transfers along the track of a fast electron referred to above which have the same LET as an electron "tail." A number of other papers are concerned with the relation between LET and the production of chromosome aberrations in plants by irradiating influorescences, pollen, or seed. The work of Swanson (336) has already been discussed (p. 47).

Kirby-Smith, Sheppard & Craig (184) have measured the RBE of fast neutrons relative to Co^{60} γ-rays with respect to the production of chromosome aberrations in *Tradescantia* microspores irradiated either in influorescences or as pollen. The RBE was estimated to be about 8 to 10 in both cases and was the same in hydrated (normal) and dehydrated influorescences. The neutrons proceeded from the internal target of a cyclotron operating at 22 Mev, but when account was taken of the degradation in the shielding, it was estimated that the neutron spectra probably had a broad maximum at about 1 Mev. Bora (36) presents extensive data on aberrations in *Tradescantia* chromosomes resulting from irradiation of microspores 72 hours before fixation. 220 kvp x-rays, 0.4 Mev Iridium γ-rays and filtered pulsed 30 Mevp x-rays were compared. At 100 to 500 r the γ- and 30 Mev x-radiations were equivalent. At higher dose levels the 30 Mev radiation had the lower RBE. Pollen lethality has been studied in Datura plants grown from seeds exposed to x-rays, thermal neutrons and fast neutrons from a nuclear detonator or from a cyclotron (the cyclotron of the Oak Ridge National Laboratory) by Spencer & Blakeslee (324). Thermal neutron doses were high enough to impair germination severely, but germination was at the 90 per cent level with the other irradiations. The RBE of fast neutrons relative to x-rays was estimated to be 13 to 14 for mutation rates (mainly chromosomal) based on pollen lethality. Cytological examination of the pollen of plants derived from irradiated seed by Cummings, Goldstein & Blakeslee (69) confirmed that neutrons were much more effective than x-rays in causing chromosomal abnormalities.

Barley seeds have been irradiated at different positions in the Norwegian heavy water reactor, and the growth rate, survival in the field, fertility, and mutation rate measured and compared with the results of exposure to known doses of x- or γ-radiation. Thermal neutrons appear more efficient per rad than fast neutrons, and were estimated to have a RBE of 10 to 40 relative to x-rays with respect to the four criteria studied. A different mutation spectrum was obtained with radiations of different LET [Ehrenberg & Saeland (90)]. In a survey paper entitled "The effects of x-rays, 2 Mev electrons, thermal neutrons and fast neutrons on dormant seeds of barley," Caldecott (49) brings together new as well as published experiments relating to chromo-

some aberrations induced by exposing dormant barley seeds to low and high LET radiations and to the viability of seedlings from the irradiated seed. The greater uniformity of response to thermal neutrons as compared with x-rays is attributed to the fact that water content, which is an uncontrolled variable, has little influence on thermal neutron sensitivity (cf. Section 6a). The fact that per unit survival, more than twice as many interchanges result from irradiating dormant seeds with thermal neutrons as result from exposure to x-rays is offered as evidence that x-rays either affect the genetic apparatus in an undetected manner or produce events of an extrachromosomal nature which are largely responsible for death of the organism.

380 Mev alpha particles, 190 Mev deuterons and 210 kv x-rays have been compared with respect to cytological damage to rabbit lens epithelium, by Von Sallmann, and his co-workers (352). The whole mount technique of Howard was employed so that all dividing and degenerating cells of the entire population of the epithelial layer could be counted, and mitotic activity was followed throughout the period of depression and the following compensatory wave. In the first place, taking advantage of the relative absence of scattering of high energy particles, selective deuteron exposure of the equatorial and central areas of the lens were made which placed on a firm experimental basis previous impressions that the sensitivity of cells in peripheral germinative zone is high in contrast to the relative radioresistance of cells of the larger central part. With respect to mitotic inhibition no significant difference was found in the RBE of these radiations. Injury to the nuclei, however, produced by equal doses of the three radiations was much greater in the case of deuterons and alpha-particles, and the latent period before clinical evidence of damage to the lens was shorter. Relative to x-rays the RBE with respect to nuclear damage was estimated at 3.4 (2.7 to 6.1) for alpha-particles and 3.9 (2.3 to 6.6) for deuterons. The latter figure is very puzzling, since the LET of the 190 Mev deuterons, as conventionally calculated, is about the same as for 200 kv x-rays and these two radiations have been shown by Tobias (340) to be equivalent as regards survival of haploid and diploid yeast. Age induced cell degeneration in the lens showed a cytopathology strikingly similar to that induced by ionizing radiation. Biegel (29) reports a clinical and histological study of the effects of a range of doses of 16 to 19 Mev electrons and 23 Mev γ-rays on the eye of the rabbit.

Leinfelder, Evans & Riley (210) who summarize some salient features of cataract induction by x-rays and neutrons considered as a "radiation hazard" emphasize that the RBE of neutrons relative to x-rays is always found to be larger for fractionated than for single exposures, and varies with neutron energy. An important influence of metabolic state on the relative biological effectiveness of α and x-radiation has been brought to light by Elkind & Beam (92) who have compared the survival of a haploid strain SC/7 of *Saccharomyces cerevisiae*, freshly harvested from a rich culture medium, with that of the same cells cultured on dextrose and salts but starved of nitrogen for up to nine days. Considering the survival of cells which have not budded

at the time of irradiation and which are exponentially inactivated by both radiations, the effect of starvation is to increase the sensitivity to α-radiation very slightly in the first 24 hours, and to decrease the x-ray sensitivity by a factor of about 2.5 over a period of about five days, with the result that the RBE of a α radiation relative to x-radiation changes from 0.6 to 2.0 over the five-day period. One is reminded of the fact that, whereas vegetative forms of bacteria have shown an RBE of α radiation relative to x-radiation which is less than unity, spores in general show values greater than unity. In the case of freshly harvested haploid yeast, budded cells have an RBE of about two for α radiation compared with a value of about 0.6 for the cells which have not commenced to bud. Since starvation progressively reduces the proportion of budded cells, a very complicated influence of starvation on sensitivity would be observed if care were not taken to distinguish between effects on budding and on nonbudding cells.

Gärtner (106) has compared mitotic activity and cell degeneration in tissue cultures of chick heart fibroblasts exposed to 184 kv x-rays, 10 Mev monoenergetic electrons and 16 Mev x-rays. Many experimental observations covering the range 250 to 2000 r are presented. The effects of 31 Mev x-rays and 180 kv x-rays on Ehrlich ascites tumour cells have been compared by Reiche (287). Animals bearing seven to nine day old tumours were exposed to 35 to 500r and mitotic figures carefully analysed as to phase, mitotic frequency and abnormality of cells withdrawn 2 to 48 hours after irradiation. No significant difference in RBE of the two radiations was found. Qualitative differences between effects of 40,000 r of 15 Mev electrons and 60 kv x-rays on mouse ascites tumour cells are reported by Hofmann (142). The β-rays of P^{32} have been estimated by Seamen *et al.* (312) to have an RBE slightly less than 0.5 relative to 200 kv x-rays with respect to the production of erythema of the skin of the rabbit. Amy (7) has reported a careful comparative study of gross and histologically observable developmental injuries produced in haploid *Habrabracon juglandis* exposed as young embryos to equivalent doses of x-, γ-, and β-radiation in the range 100 to 350 rad. No qualitative differences between these radiations were noted. As regards hatchability x-radiation was clearly more effective than γ- or β-radiation (of the order of 1.5:1). Differences between β- and γ-radiation were perhaps doubtfully significant, when all errors of measurement were allowed for. The three radiations had about the same relative effectiveness as regards adverse effects occurring in the larval stage. Pupation without cocoon formation was a frequent occurrence and positively correlated with dose. Heidenthal, Clark & Gowen (138) report an RBE of unity for 50 Mev x-rays compared with 122 kv x-rays with respect to induction of dominant lethals in *Habrabracon* larvae irradiated as eggs in meiotic metaphase.

The effects of 180 kv x-rays and 31 Mev x-rays have been compared with respect to effects on bristle development resulting from irradiation of the five hour old *Drosophila cryhsalis* [Naville (246)]. The biological significance of natural background radiation at sea level and at high altitudes is discussed

by Schaefer (308), taking into account statistical variations in the energy absorbed in cells exposed to particles of different LET.

8. The Influence of Fractionation

Lamarque & Gary-Bobo (198) have studied fractionated x-irradiation of eggs of the insect Bacillus rosii. After a single dose survival is sigmoidal and half the eggs hatch after 1250r. The effectiveness of two equal fractions progressively decreases as the interval is increased to 24 hours, but sensitivity then progressively increases with further extension of the interval to two to four days. Elsewhere (199) these authors integrate the observations referred to above with others on recovery exhibited by the eggs of *Bombyx mori*, stored at different temperatures, and recovery of human skin. On the basis of a small series of observations, Healmke (140) reports an effect of fractionation on ultraviolet inactivation of *E. Coli.*

9. Radiation Carcinogenesis

Currently there is much interest in the development of leukaemia as a result of whole body or partial body irradiation, in cancer associated with hormonal imbalance resulting from destruction of a gland by irradiation, and cancer arising from injection of radioactive material (c.f. section *Carcinogenesis and Metabolism of Bone-Seeking Isotopes*—U.N. Report pp. 134–169) but few studies at the cellular level have been reported. The whole subject has been well viewed by Brues (43) with comments by Kaplan (165). Rogers (296) has observed that when small fragments of foetal lung are exposed *in vitro* to a single dose (1100 micro-watts per square centimetre for 15 minutes) of 2537 Å ultraviolet radiation and reimplanted intramuscularly into mice of the strain of origin, pulmonary adenomas developed. This did not occur with unirradiated but similarly treated material. Analogous results have been obtained by the same author by nitrogen mustard treatment. The carcinogenic effects of methyl clolanthrene and β-radiation of ^{32}P or ^{91}Y applied locally to the skin of mice have been observed by Cloudman *et al.* (58) to be approximately additive when a given area of skin is treated by both chemical and ionizing agents. There was a suggestion of some synergism between them in the production of papillomas. Van der Scheuren (346) reports an interesting series of observations on biological damage sustained by frogs exposed once or repeatedly to ultraviolet radiation. The investigation follows up the author's observations on the carcinogenic action of ultraviolet radiation in mice. The observations on frogs include studies of survival, movement of potassium and sodium in each direction through the skin, sulphonamide protection and histological aspects of skin damage. Big species differences between the green and brown frogs *Rana esculenta* and *Rana temporaria* were observed, the latter being much more easily killed by whole body ultraviolet exposure. This aspect of ultraviolet damage was photoreactivable, and temperature dependent.

10. The Mutational Process and Genetic Damage

Soon after Müller first demonstrated that ionizing radiation could induce mutations in *D. melanogaster*, evidence was obtained which was taken to indicate that the mutational step could be reversed by a subsequent irradiation. The experimental grounds for believing that such reversions occur has been questioned by Lefevre (209) and the conclusion has been drawn that in *Drosophila* and maize ionizing radiations produce, in addition to chromosomal rearrangements, only destructive gene mutations. Giles, de Serres & Partridge (113) have now adduced rather convincing evidence that true reverse mutations may in fact be induced in *Neurospora crassa* at the inositol locus, and probably also at the purple-adenine locus by exposure to ionizing radiation. *Neurospora* offers many technical advantages for such studies including the fact that reversion frequencies of the order of one per 10^7 surviving conidia may be evaluated. The main part of the work is devoted to a study of x-ray induced reversions of mutations originally induced by ultraviolet or by chemicals. X-rays are found to induce dominant positive mutations resulting in the restoration of specific biochemical syntheses of which a minority are due to the independent induction of a suppressor mutation and the majority behave, as far as could be ascertained by genetical analysis, as true reverse mutations. Some preliminary studies have been made of the reversibility by x-rays of x-ray induced purple-adenineless mutants. Five of the original mutants showed irregular genetic behaviour and were not induced to revert by x-rays. Four behaved normally, and of these three have undergone reversion either spontaneously or following irradiation. It is concluded that in *Neurospora* not all x-ray induced forward mutations are destructive, and that x-rays are capable of inducing positive dominant changes, some of which are most simply interpreted as true reversions. The experiments of Hollaender & Stapleton (154) on the influence of cysteamine pre-treatment and post-irradiation culture conditions on the yield of auxotrophs recovered after irradiation of an adenineless strain of *E. Coli* B/r have already been referred to. In this case the mechanism whereby the organisms regain the ability to grow without an exogenous supply of adenine has not been established. Whittinghill (358) has made a detailed study of x-ray induced crossing over in the *Drosophila* male where it does not occur spontaneously, and of increased rate of crossing over in the irradiated female. No evidence of departure from a strictly linear relation between x-ray dose to mature sperm of the wasp *Mormoniella vitripennis* and the number of recessive lethals linked to a marker gene has been observed by Saul (305). The rates for two groups of recessive lethals linked to different markers were 0.004 per cent and 0.001 per cent per r respectively. Lethal damage could be expressed at any stage between egg and adult. Sommermeyer (323) discusses the production of mutations by irradiation of *Drosophila* sperm in terms of classical target theory, but with mutation rates 5 per cent times higher than were formerly taken, to allow for mutations which affect viability. He finds considerations relating to spread of energy from the point of primary

absorption to be superfluous, and identifies the volume which must be directly ionized with the volume of the chromomeres. Richter & Singleton (290) have shown that chronic exposure of carnations to γ-radiation at dose rates of between 1 and 200r per day results in the production of somatic mutations which can be vegetatively propagated. Breider & Reichardt (39) have produced sectorial somatic mutants by irradiating dormant or sprouting buds of vine and embryos of cyclamen, and have described interesting features of the mutants. The broad aspects of radiogenetics are reviewed in a subsequent paper [Breider (40)]. Rowell (302) has observed that exposures of vegetative cells of a diploid line of corn smut fungus *Ustilago zeae* to γ-radiation induces an unstable condition that results in segregation and recombination of the factors for sexual compatability during multiplication of the affected cell. Price (280) has examined interspecific hybridisation between normal and irradiated games of Secale. By exposing the moss *Bracythecium rutabulum* to x-radiation at meiosis 7 new strains were obtained which cytological observations showed were all aneuploids having chromosome numbers ranging from 10-5. [Moutschen (239)]. Emmerling (93) has made a cytological analysis of ultraviolet and x-ray induced genetic effects produced on chromosomes 9 and 10 of Zea mays. Quantitative differences were found, in the sense that a lower frequency of complex deficiencies were produced by the ultraviolet. In a short paper Gustafsson (130) draws upon the extensive research carried out over many years by himself and his colleagues at the Swedish Forest Research Institute (131) to show how the proportions of mutations of different kinds in barley may to some extent be controlled by the choice of a suitable mutagen, ionizing radiation of high or low LET or a radiomimetic chemical. He points out the importance of this from an agricultural point of view, since a few mutants in the heterozygous condition are directly beneficial to the organism, augmenting viability both as regards vegetative matter and seed production even under conditions optimum to the mother strain. *The Proceedings of the Geneva Conference* contain an important group of four papers by Muller (243), Russell (304b), Wallace (353) and Carter (54), in which experimental data for induced mutation rates in *Drosophila* and mice are presented, and the question of mutations in natural populations are discussed in relation to the problem of the genetic hazard to a human population exposed to ionizing radiation.

Müller (240, 241, 242) has attached great importance to subvital mutations in natural populations exposed to ionizing radiations. Falk (97) throws light on this question by observing the lower hatching rate of eggs of progeny of irradiated flies in which, by a special technique, reduced viability due to mutations in the third chromosome of *D. melanogaster*, are isolated for study. At a dose level of 2000r about three-fourths of the third chromosomes are so damaged as to affect viability, and subvitals were found to be at least 3.5 times as frequent as lethals. It was concluded that dominant mutations are very frequent among subvitals. Arising out of a statement by Cockcroft (59) concerning radiological hazards from nuclear explosions, Haldane (134) con-

tributes some considerations of possible levels of genetic damage arising from products of such explosions and Cockcroft (60) comments on Haldane's estimates. A status report of information on the induction of mutations by radiation, and some assessments in relation to man and the radioactivity of his environment, have been made by Slatis (320). Papers dealing with various aspects of radiation genetics are discussed elsewhere. [Cf. references on effects of radiation on chromosomes (36, 37, 49, 51, 61, 63, 64, 68, 76, 77, 88, 93, 125, 132, 135, 155, 174, 175, 194, 200, 209, 220, 288, 293, 306, 307, 335, 336, 362, 363).]

II. RADIOBIOLOGY OF CELLS AND TISSUES

1. Bacteria and Thymocytes

Effects of radiations of all kinds, from the short visible region of the spectrum to high energy ionizing radiation, on bacteria are excellently reviewed by Zelle & Hollaender in Chapter 10 of *Radiation Biology* with a bibliography of over 200 references to original papers, mainly prior to 1953. Currently Stapleton (325, 327) discusses factors which influence bacterial inactivation by ionizing radiation and has described the manner in which the sensitivity of *E. Coli* B/r varies throughout the division cycle, Biagani (28) has made measurements of the inactivation of *E. Coli* B at different concentrations which led him to conclude that some of the inactivation was due to effects of radiation on the suspension medium, but it would appear that such interpretations are open to doubt because no mention is made of any measurement or control of the oxygen tension in the medium during the rather prolonged irradiations. Böttcher (38) has studied the inactivation of *E. Coli* by 60 to 180 kv x-rays from the standpoint of biological dosimetry. A large literature on the bactericidal action of ionizing radiation has arisen out of interest in the application of this different radiation to the preservation of food and pharmaceuticals. Currently Proctor, *et al.* (281) have studied the inactivation of *B. subtilis* under environmental conditions. The whole field has been reviewed by Bellamy and co-workers (25).

Schrek (309) makes a further contribution to the radiation induced degeneration of lymphocytes by means of time-lapse cinemiographic observations of rabbit thymus lymphocytes degenerating after exposures of 1000r delivered *in vitro* at 37°C. The actual process of degeneration appears remarkably similar to that which occurs naturally and to take about the same length of time, but the onset of degeneration occurs much earlier in the irradiated material. Interesting aspects of the radiation induced degeneration of lymphocytes, irradiated *in vitro* have been discussed by Patt (259). [Cf. Section I (8, 26, 82, 111, 121, 133, 139, 140, 153, 201, 203, 205, 224, 225, 260, 276, 279, 281, 326, 329).]

2. The Cellular Elements of Testis and Ovary

The effect of x-radiation on the cellular elements of the mouse testis has been studied in great detail by Oakberg (250 and 251). In this organ [by

contrast with the ovary, studied by Deringer, Lorenz & Uphoff (74)], changes in weight and histological appearance after doses of 60 to 210r have been shown by Kohn (188) to be a local effect of radiation which (in the rat) is not sensibly affected by irradiation of the rest of the body. Moreover, Kohn & Kallman (190) have observed, in agreement with earlier observations, that weight loss in mouse testis four weeks after exposure to x-radiation is independent of fractionation over fairly wide limits and proportional to dose, indicating a monotopic type of primary damage.

Unlike some previous authors, Oakberg (251) concludes that reduction in the number of spermagonia after irradiation can be explained by killing of cells without postulating a prolonged mitotic inhibition of spermatogonia. One recognisable class of spermatogonia appears in significantly smaller numbers at metaphase or anaphase after as little as 20r. This is ascribed to breakdown in late interphase or on entering division. At the other extreme spermatids and sperm show no visible changes after doses up to 1500r though they are known, of course, to carry genetic damage. Spermatogenic cells in the mouse show qualitative and quantitative differences in sensitivity that are co-related with the dramatic nuclear changes in spermatogenesis, and the author submits that the primary reactions leading to the degeneration of these cells are nuclear. The effects of long continued small daily doses of x-rays on spermatogenesis in dogs has been studied by Casarett & Hursh (55). No effects of radiation were conclusively demonstrated at 0.3r/week or 0.6r/week over a two year period but there was a progressive decline in sperm count in dogs receiving 3r/week. It is interesting to compare the observations of Oakberg with those of Alexander & Stone (2), who studied the induction of dominant lethals and of translocations in germ cells of *Drosophila virilis* as a function of cell development through eight spermatagonial stages to mature sperm. A late spermatagonial stage immediately preceding meiosis is evidently very sensitive as regards immediate cell lethality and may correspond with the very sensitive stage observed by Oakberg. Irradiation at this stage yields a few sperm, a high proportion of which carry dominant lethals, but extremely few translocations. Most recoverable translocations arise from the irradiation of immediately post-meiotic cells. They observed that anoxia greatly reduced the yield of translocations derived from irradiated pupae, in agreement with Baker and Edington (18), and also the yield of dominant lethals. Baker and Von Halle (20) found only a small effect of anoxia on dominant lethals resulting from irradiation of the male fly. An appreciably higher proportion of both translocations and dominant lethals was observed after exposure of pupae to a given dose in carbon monoxide than in nitrogen, which the authors discuss in biochemical terms. Experience shows that it is often difficult to render tissues anoxic simply by changing the gaseous atmosphere in the absence of respiration. Since the irradiations in the present investigation were carried out at 1°–3°C. and since Baker & Edington have shown that translocations frequently rise steeply with oxygen tension, it may be that the effect of the carbon monoxide was to prevent com-

plete anoxia being attained. Glass (114, 115) has taken up the comparatively neglected question of the mutation rate which results from irradiation of the female germ cells of *Drosophila*. One paper is devoted to a study of translocations and inversions, and the following paper to deficiencies and minutes. Oöcytes were irradiated in first meiotic metaphase and sperm from the same stock of flies were irradiated at the same time with the same dose, so as to secure as close as possible a comparison between the response of the two gametes. The most striking results of the first investigation were the extremely low frequency of translocations recovered from irradiated oöcytes, namely one in 2599 oöcytes, compared with 150 in 2387 spermatozoa. Thinking that this might possibly be due to a low probability of rejoining, rather than low breakage rate, infrared was used as pre- and post-treatment to supply conditions favourable to rejoining. The infrared was applied at reduced temperature and in all respects followed as far as possible techniques reported to be effective by previous workers, but no effect of pretreatment, and little, if any, effect of post-treatment, was observed even in the case of the sperm. Glass used genetic tests for translocation, since this permitted detection of numerous translocations between heterochromation which are very difficult to detect cytologically. The effects of infrared were therefore tested by a procedure which required full development of the fly, whereas cytological examination only requires development to a larval stage. Such a complete divergence of results can, however, only be clarified by further experiments. More x-ray induced translocations were observed in the second brood (five to seven days) than the first (one to four days) in agreement with other workers. In the *Drosophila* oöcyte, as in *Sciara* and *Habrabracon*, sensitivity is apparently minimal until meiotic prophase begins. In the second paper by contrast, sperm and mature oöcytes are found to have almost exactly the same sensitivity with respect to induction of "minutes." Inversions were less frequent in oöcytes than in sperm, but the disparity was not nearly as great as with translocations. A doubling of frequency of minutes occurs in the third brood (eight to eleven days) from treated males, and a great drop is observed in the third brood from treated females. It is pointed out that most minutes may be identified as small deficiencies, which occur with equal frequency in male and female gamete, and are linearly related to dose at low doses, indicating that they arise as a result of a monotopic (one-hit) process. It is argued therefore that the chromosomes are broken with equal frequency by a given dose in whichever gamete they occur, but that the probability of reunion between broken ends decreases with distance between the broken ends more rapidly in the oöcytes than in the spermatozoa. Discussion is to be continued in a later paper. King & Wood (182) have measured the frequency of sex-linked recessive lethals resulting from thermal neutron irradiation of *Drosophila* gametes, both sperm and oöcytes. All observed mutation rates varied linearly with dose. The freuqency of mutations recovered from irradiated sperm was constant for matings from day 1–10, but when oöcytes were irradiated, the

frequency fell from 75 per cent of the value for sperm in 0–6 day eggs, to 52 per cent in 6–8 day eggs, and 22 per cent in 8–10 day eggs. The question was raised as to whether these differences might be due to different oxygen tension in the male and female gonad. This would seem a little improbable in the case of a pure thermal neutron effect, operating through the reaction $N^{14}(np)C^{14}$. However, in these experiments it was estimated that 430 rep of 5 Mev γ-rays contaminated the thermal neutron column and were added to the 10^{13} thermal neutron/cm.^{2} The two radiations together gave a mutation frequency in sperm equal to that produced by 740r of x-rays. Since increasing the Li and B contents of *Drosophila* tissues by 2 and 100 respectively did not change the effectiveness of the thermal neutron irradiation it was considered [King (179)] that the only heavy particles giving rise to mutation were protons from $N^{14}(np)C^{14}$, which contributed 293 rep per 10^{13} n_{th};/cm.^{2} and C recoils, giving 23 rep per 10^{13} n_{th};/cm.^{2}. On this basis the heavy particles would appear to have an RBE of 1.5 relative to x-rays with respect to induction of recessive lethals in sperm. King (180) has also examined the production of dominant lethal mutations and x chromosome eliminations after x-irradiation of female *D. melanogaster* as a contribution to the problem raised by the decreasing yield of genetically damaged eggs in successive batches laid by the irradiated female. Oster (255) finds that *Drosophila* spermatagonia, in the stage of development reached in 48 hour pupae, to be more sensitive to the induction by x-rays of translocations between chromosomes 2 and 3 than older pupae. Moreover, the ends of chromosomes broken during spermatogenesis are found to undergo reunion before fertilisation, while those produced in mature spermatazoa remain open and reunite during fertilisation. Observations substantially in agreement with those outlined above on the relative sensitivity of different stages of spermiogenesis in *Drosophila* have been reported by Fritz-Niggli (102) for irradiation with both 180 kv and 31 Mev x-rays. The very much greater sensitivity, as judged by hatchability, of oöcytes of *Habrabracon* when exposed to x-rays in meiotic metaphase, as compared with cells exposed at prophase I, has been shown by Von Borstel (350) to be true also of treatment with nitrogen mustard. Dose response relations are also strikingly similar. However, 100 per cent lethal doses of nitrogen mustard induce mainly single breaks, whereas x-rays induce isochromatid breaks. In a study of the induced mutation rate in *Drosophila* flies which were grown from the first instar larval stage onwards in food containing ^{32}P, Bateman (23) points up the many difficulties associated with a comparison between ^{32}P and x-ray induced mutation rates. In agreement with x-ray studies (2) it was found that there is a hypersensitive stage of spermeogenesis which was estimated to occur seven days prior to maturity. Since infrared pre- and post-treatments are known to influence the frequency of breaks induced by ionizing radiation [c.f., however, Glass (114, 115)], it is of interest to note that 3 hours treatment with 1μ infrared radiation alone at 20°C. applied at meiotic prophase has been shown to change the proportion of interstitial and terminal chiasmata in

Tradescantia bracteata, possibly by affecting protoplasmic viscosity [Snoad (321)]. The effect of large doses of *in vitro* radiation on the motility of human sperm has been studied by Gerber (107). [Cf. Section I (256).]

3. The Developing Embryo

In the five years since Russell reported the extensive damage sustained by the mammalian embryo by as little as 100r of x-radiation delivered during a comparatively short period of organogenesis, much information has accumulated. It is briefly reviewed by Russell & Russell (304a). Currently Rugh & Wolff (304) report that though very extensively damaged by four hours after exposure to 300r, the eye of the 13 day mouse foetus has such powers of reconstruction from more radioresistant precursor cells that the eye may appear essentially normal at birth six to seven days later. Brunst (45) reports a detailed histological study of the influence of x-radiation on the development of the eye of the axolotle. The sensitivity of some retinal elements is emphasized. The effects of x-rays on the embryonic development of the snail have been followed in detail by Bonham (33) from first cleavage onwards. By the criterion of survival to hatching, snail eggs at metaphase or anaphase of early cleavage division are most vulnerable, and then have a sensitivity comparable with that of *Drosophila* eggs. As is the case with *Drosophila* eggs, sensitivity falls with embryonic development. A preliminary investigation has been made by Wellaender (354) with trout eggs. The effects of x-radiation on the mitotic index and on the appearance of degenerating cells in *Xenopus* larvae has been studied by Grossmann (129) and on regenerating tails of *Xenopus* larvae by Höhn (151). Rostand (300) describes a variety of abnormalities in frogs (*Rana temporaria*) derived from the 20 per cent of larvae which survived a dose of ultraviolet radiation. The neuroblasts of the grasshopper embryo are outstandingly radiosensitive as regards mitotic arrest, more particularly cells in late prophase. The x-ray sensitivity of these cells with respect to the induction of chromosome stickiness also depends very much on the phase of the cell at the time of irradiation. Sensitivity sharply decreases in passing from late prophase towards anaphase [Carlson & Harrington (53) cf. Section I (7, 39, 51, 56, 66, 122, 161, 191, 257, 263, 337, 338, 341, 349, 357)].

4. Skin and Vascular System

Devik (78), in a monograph entitled "A study of the local röentgen reaction on the skin of mice, with special reference to the vascular effects," reviews the literature and contributes many new and interesting observations By stretching the loose skin on the back of a "hairless" strain of mice into a semi-circular bow, blood vessels may be examined by transillumination, and the spread of fluorescent dyes studied. In this way vascular changes were followed continuously after administration of a standard dose of 2700r of 50kv x-radiation, and *in vivo* appearances compared with the histological appearance of frozen sections. It is concluded that the acute skin reaction is probably

mainly dependent on radiation effects on the epithelial cells. The vascular factor is considered as being only of secondary importance in the pathogenesis of the acute skin reaction. A regular and very interesting feature of the observations was the appearance of a hyperplasia within the irradiated area at the boundary of a sharply defined irradiation field. When small pencils of radiation 1 to 3 mm. in diameter were used, the whole irradiation field might be occupied by markedly hyperplastic epithelium six days after exposure to the standard dose or even to the very much higher dose of 15,000r. This observations emphasizes the great extent to which the fate of irradiated cells depends on the proximity of healthy tissue. By a plastic window technique Merwin & Hill (229) have observed directly the rate of formation of new capillaries in a lightly burned region of mouse skin or around stitches of tantalum wire in control and locally irradiated (body shielded) mice. At about the 1000r level upwards irradiation markedly delayed revascularisation. Some rather immediate effects of local irradiation on tissue have been inferred from the greater spreading of India ink and of diphtheria toxin injected under an area of rabbit skin exposed to doses of from 250 to 2000r [Prodi & Miceli (282)]. Direct measurement of the fragility of the capillaries of the mesentery of the frog by insertion of a 2–5μ micro-pipette connected with a manometer has shown a very significant increase in fragility by the time of onset of irradiation sickness in animals totally exposed to 3600r. From comparative observations with heparin, tripsin and peptone it is believed that heparin is implicated in the radiation response [Rieser (292)]. Changes in the blood vessels of the rabbit, based on perfusion tests, 3 to 37 days after exposure to 10,000r, have been studied by Hate & Schrader (136). In order to isolate the influence of temperature per se on skin reaction from effects due to simultaneous changes in tissue oxygen tension, O'Brien, Frank & Garner (252) have exposed the ears of white mice to 29,000r of x-rays at 10°C. and 39°C. while the skin was held anoxic by compression. Reactions at all times from 24 hours to 8 weeks were greater in skin irradiated at the higher temperature. Joyet & Hohl (163) find that as regards the influence of field sizes on skin reaction to radiation delivered as in deep x-ray therapy with standard fractionations, a large number of observations can be represented by the simple relation that the surface dose which produces a given skin reaction varies linearly with the reciprocal of the irradiated area. Exposure of tail epithelium of tadpoles to 2000 to 4000r leads to formation of giant cells, often multinucleate. The formation of giant cells has often been reported to result from the irradiation of plants, insects, amphibia, and the normal and malignant cells of mammals. These giant cells appear to be in the process of degeneration in the tail epithelium of the tadpole and even more prominently in irradiated regenerating tails. The degenerate giant cell epithelium is very frequently bordered by a strip of epithelium at a high level of mitotic activity which gives rise to outgrowths. This stimulated growth is ascribed to the products of cellular disintegration [Brunst (44)]. The belief that when a salamander limb regenerates after amputation a large part

of the limb arises from epidermal cells, has been tested by Rose, Quastler & Rose (297) by allowing the remainder of a heavily irradiated limb, itself incapable of regeneration, to be covered after amputation by unirradiated skin. Hands regenerated which, though smaller, were complete even when grown on a stump which had received 10,000r. This was considered to establish the fact that an entire limb may regenerate from epithelial cells. [Cf. Section I (58, 103, 128, 143, 144, 146, 199, 217, 312, 346, 352).]

5. Thyroid and Orbital Glands

A recent study by Doniach & Logothetopoulos (83) of thyroid damage in rats resulting from the administration of large doses of I^{131} (30μC per 60 mgm. thyroid) brings out very clearly the long period over which nuclear damage induced by ionizing radiation may be stored in the absence of cell division. The interphase time is extremely long in the cells lining the follicles, and correspondingly response to hemithyroidectomy or a goitrogen given as late as three months after irradiation, at a time when the thyroid, though smaller than normal, was fully functional, results in appearance of bizarre mitotic figures and micronucleic characteristic of chromosome structural damage. It is interesting to reflect that in this typically "radioresistant" tissue the integrated amount of cell degeneration, which occurs normally so slowly as to escape detection, may be as great as in many sensitive tissues. Interesting light is also thrown on the delay of radiobiological damage by the experiments of Maloof (223), who has observed that cellular hypertrophy and nuclear damage in rat thyroids which had been heavily irradiated with I^{131} is temporarily suppressed by hypophysectomy or administration of DL-thyroxine but appears two months after the cessation of thyroxine treatment. The destructive effects of I^{131} on transplants of rat thyroid carcinoma tissue have been studied under conditions related as closely as possible to the clinical use of I^{131} in therapy [Forbes (100)]. As suspected, cell destruction is limited to regions containing well differentiated cells where differential uptake is high. Thyroid destructoin in the dog has been studied by Levene, Andrews & Kniseley (211). Histological changes in human thyroid tumours following I^{131} therapy have been reported by Miller, Lindsay & Dailey (232). Cogan, Fink & Donaldson (61) report on the histological damage to the orbital glands of rabbits which have received 1000 to 4000r of x-radiation.

6. Nerve, Retina and Muscle

With the publication of the papers presented at the Geneva Conference many English speaking radiobiologists have become aware for the first time of the trends of work in the U.S.S.R., and it is interesting to note the attention which has been given to the effects of radiation on the nervous system. Lebedinsky (207) reports at some length on reactions of the nervous system in totally irradiated animals. Of recent years there appears to have been a general revival of interest in this branch of radiobiology, which holds promise of a better understanding of the effects of radiation on fully differentiated

cells. Bering, *et al.* (27) review the subject from 1896, when Tarkhanov noted that reflexes became slowed in dogs, and flies became sluggish after exposure to x-rays, through 1934, when Nemenov reported that doses of 1500r and over would produce a state of inhibition of conditioned reflexes in dogs, to 1943, when Warren, surveying the field, concluded that peripheral nerves and the autonomic system were highly resistant to radiation. This is a view which generally prevails today. This subject, perhaps more than most, lends itself to inconclusive experimenting, but it is also possible that criteria of sensitivity have not been adequate, having regard to the nature of the cell under investigation. In the case of *in vivo* irradiations it will be difficult to separate effects on the nerve cells and the neurons from effects secondary to changes in surrounding tissues. Bering *et al.* (27) have introduced ^{182}Ta wire into the brains of monkeys as a source of local radiation. The wire remained in position until the animals were sacrificed 18 to 20 days later. From clinical and histological observations it was concluded that the changes were more extensive than could be accounted for by vascular damage alone, and direct neuronal and glial effects were considered to be indicated. Using the same technique to irradiate the spinal cord these authors observed that paralysis only set in, and spinal electrograms were only markedly deadened, by doses of upwards of 5000r in a single day. The cord showed great powers of recovery and fractionation considerably increased the tolerable dose [McLaurin *et al.* (219)]. Commenting on this work Krabbenhoft (192) directs attention particularly to the experimental work of Arnold (10, 11, 12, 13) on the effects of radiation on the primate brain. Local irradiation (6000r) of the brain of the anaesthetised cat with 215 kv unfiltered x-radiation at 50 to 135 r/min. has been reported by Lee, Helgason & Snider (208) to diminish cerebellar response to the stimulation of peripheral nerves. This deadening of the response to nerve impulses, which was observed during irradiation, was more marked when the dose was increased to 15,000r. The cerebellar response showed a further reduction during a 50 minute period after irradiation. The basic neural mechanism which is affected by x-rays in such a way as to block conduction is not known and the authors expressly include vascular damage as a possible contributory cause. When sciatic nerves of bull frogs are irradiated *in vitro* at 9000r per minute, doses in excess of 75,000r cause a decrease in conduction velocity, and doses above 300,000r produce complete abolition of nerve conduction within one hour following irradiation. Rabbit nerves, however, proved to be much more radiosensitive than frog nerves, since doses in excess of 45,000r produced a complete block within one hour [Gerstner, Orth & Rickey (109)]. 12,500r of x-rays delivered in 24 minutes to the head of the rabbit is followed after a latent period of half an hour by a convulsive phase, a somnolent phase, and finally by an ataxic phase affecting posture and movement until death on the second or third day [Gerstner, Pickering & Dugi (110)].

Lipetz (213) reports an extremely interesting quantitative study of retinal response to x-radiation. The frog's retina was surgically exposed *in vivo* to

eliminate effects due to x-ray induced fluorescence of the media of the eye other than the retina. The character of the electroretinogram under repeated x-ray stimulation was qualitatively similar to that for visible light. Threshold doses were of the order of 1 roentgen, e.g. 0.2r/sec. for 5 secs. and were estimated to correspond to from one-half to 160 times as much energy absorbed by the visible purple as the minimum for visible light stimulation. The dose needed to bleach the visual purple of the dark adapted retina was 10^7r, which caused protein coagulation and some decomposition. This exceeded the threshold dose for stimulation (less than 1r) by a factor of the same order of magnitude as the factor of 10^7–2.10^8 by which the amount of visual light needed to bleach the visual purple exceeds the threshold for stimulation. It was observed that a threshold stimulation for x-rays lowered the sensitivity of the retina to x-rays about five times as much as a threshold light stimulation lowered responsivity to light, and that frequent near-threshold x-irradiation produced a cumulative rise in light threshold, though light had no such marked effect. Moreover, even these small doses of x-radiation temporarily reduced the voltage of the ganglion cell discharges, which light did not. It was concluded that x-rays have an action on the dark adapted retina essentially similar to that of light, but in addition have a desensitizing effect. The effects of ^{60}Co γ-rays on the retina [Kalaschnikov (164)] and on the nerve elements of the cornea [Krestinskaya (193)] of the rabbit have been studied.

The pattern of clinical and histological changes which develop in the retina of the monkey following administration of large doses (10,000r) of ^{60}Co. γ-rays has been followed in detail over the first 96 hours by Brown, Cibis & Pickering (42). Histological abnormalities consist chiefly of pyknosis of the rod nuclei and degenerative sequelae in the outer nuclear and bacillary layers. Wilde & Sheppard (359) find that intense local irradiation of muscle (58 kr) leaves muscle still functional for up to 22 hours. Marked extracellular oedemia is produced, but the fibres lose no potassium per unit dry weight. [Cf. Section I (156, 247)]

7. Animal Tumours

When the behaviour of tumour transplants under irradiation is being assessed it is of importance to give due weight to factors other than radiobiological damage to tumour and stroma which are generally involved on account of the genetic differences between tumour and host. Only when the greatest care is exercised to secure genetic identity may immunological factors be disregarded [Goldfeder (117)]. Cohen & Cohen (62), following up earlier work on the radiosensitivity of implanted tumours differing genetically in a controlled manner from the host, have found that as early as the first passage of a C3H mouse mammary carcinoma through C3H mice a spontaneous mutation occurred which significantly affected radiosensitivity as well as growth rate.

In the course of a study of the growth in the peritoneal cavity of Ehrlich

ascites tumour cells which had been exposed to 1100r of x-rays under anoxic conditions *in vitro*, Revesz (289) has observed that tumour cells damaged by 5000r and inoculated at the same time as less irradiated cells may exert either an inhibiting or enhancing effect on the growth of the viable cells, dependent on their quantitative proportion. When in excess the irradiated cells inhibited the growth of a small proportion of nonirradiated cells, inoculated simultaneously, and also prevented the development of subsequently injected ascites tumour. This was not unexpected and could be ascribed to immunisation of the host by dead or dying cells. A minority of irradiated cells, however, appeared to exert an enhancing influence on the growth of a large population of unirradiated cells, which suggested either that dying cells might release substances capable of stimulating tumour growth or that surviving cells which recovered from the effects of irradiation might later have contributed to the total number of growing tumour cells.

Gopal Ayengar (118) has compared the cytological changes resulting from exposure of Ehrlich ascites tumour cells *in vivo* to 1400r with changes produced by many different radiomimetic drugs. Variants of the Yoshida rat ascites tumour resistant to all of a group of alkylating agents of the nitrogen mustard class have been produced by Hirono (141) by passing the cells through animals treated with one particular radiomimetic. Such cells, which were also resistant to another radiomimetic chemical but not to 8-azaguanine or colchicine were tested with x-radiation. When the original strain of cells and the resistant sublines were exposed *in vivo* by irradiation of the rat to doses of 200r to 1000r and the mitotic index evaluated at intervals up to 48 hours, no significant differences were observed. From this it was concluded that the mode of action of the chemical and ionizing agents was different. The proportion of abnormal mitoses might perhaps have been a more relevant criterion of radio resistance than the mitotic index itself since the resistant strain had been produced by selection on the basis of viability in the presence of a radiomimetic agent.

Pre-transplantation irradiation with a dose of 1500r delivered locally to the leg of a mouse 48 hours before subcutaneous inoculation of a tumour cell suspension has been found to inhibit the development and growth rate of a mammary carcinoma in an experiment with adequately large numbers of treated and control animals [Stenstrom *et al.* (331)]. Evidence that the effects of whole body radiation may influence the response of the Walker rat carcinoma to radiation is presented by Hoffman & Matscke (150). 10r whole body irradiation of male albino mice is reported to produce marked changes in spleen and thymus and to interfere with the rate of growth of Ehrlich ascites tumour cells inoculated into the peritoneum [Falk (96)]. Allen (3) draws upon some experiments of his own and an extensive review of the literature in a discussion of the influence of hypothermia on radiosensitivity and on the radiocurability of tumours. Elevated temperature and environment are considered in a second paper [Allen (4)]. Schubert (310) reports briefly on radiobiological aspects of radioresistance as encountered in radio-

therapy, and Lorenz (214, 215, 216) exhaustively reviews the implications of radiobiology for radiotherapy. [Cf. Section I (63, 64, 105, 142, 158, 159, 169, 185, 195, 204, 233, 287)]

8. Germination

So many reports of a stimulating effect of ionizing radiation have not survived critical examination that a particularly cautious attitude towards such reports has been engendered. The deleterious effects of radiation are manifest. An ionizing radiation, however, excites many more molecules than it ionizes, so that a stimulating effect for any process which may be stimulated by light is not inherently improbable if circumstances are such that the deleterious effects do not overshadow stimulation. Visual perception is one such process which may apparently (cf. 213) be stimulated by x-rays as well as light. Germination may be stimulated by light and by chemical agents, and evidence is steadily accumulating in support of the view that irradiation can also stimulate the onset of germination. Ehrenberg's (88) observations hardly leave room to doubt that the effect occurs when barley seed is exposed either to x-rays or fast neutrons. Woodward & Clark (364) report a stimulating effect of low doses (500 to 5000r) of x-rays on neurospora conidia in the sense of a small increase in the number of conidia which germinate and an accelerated growth rate after irradiation. This was not observed after exposure to ultraviolet or thermal neutrons. [Cf. Section I (1, 31, 87, 132, 324)]

9. Unclassified

Although principally of interest in connection with effects observed in totally irradiated animals, which are outside the scope of this survey, mention may be made of a study of the radiation syndrome in male and female *Drosophila* melanogaster flies exposed to 70,000r. Sex differences in mortality, weight loss, and modified phosphorus turnover were noted. Oxygen consumption was not affected until the sixth day and phosphorus turnover was little affected in the male, facts which show the great radio resistance of some of the cellular activities of the adult fly [King & Wilson (181)]. Gross and cytological changes in many organs of the mosquito subjected to ultraviolet irradiation are described by Seiler (313). Inversion of the sense of the galvanotropism of a Planarian under the action of polarised visible light and ultraviolet has been observed by Viaud (348). Evidence has been sought of the influence of cosmic ray showers on the chromatophores of the fiddler crab [Brown, Bennett & Ralph (41)].

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VERTEBRATE RADIOBIOLOGY: EMBRYOLOGY[1]

JOHN P. O'BRIEN[2,3]

Department of Biology, Marquette University, Milwaukee, Wisconsin

INTRODUCTION

The importance of furthering our knowledge of the effects of ionizing radiations on vertebrate embryonic development under experimentally-controlled conditions, particularly at this point in history, is probably so obvious as to render any insistence superfluous. The interest attaching to such studies frequently and understandably derives from their probable implications for human development. Vital as such practical considerations are, they must not be allowed to overshadow the importance of radiodevelopmental investigations directed to the solution of basic problems of radiobiological and morphogenetic mechanisms, and general biology. We are frequently and advantageously reminded that when applied in the form of ionizing radiation, an incredibly small amount of energy is sufficient, ultimately, to destroy an organism; for example, 1000 r applied to the whole body is easily sufficient to kill most any mammal. This amount of radiation has been calculated to be the equivalent of only 2×10^{-3} cal. per gm. Further, it is both revealing and disconcerting to recognize, for example, that a mere 2.5 per cent of the above dose of radiation, rightly (or wrongly!) timed, is sufficient to alter, in a serious way, important features of mammalian embryonic development. Little wonder, then, that a high priority is being given to investigations into the effects of high energy radiations on vertebrate embryonic development.

It seems important to submit, at the outset, that radiobiological investigations involving ionizing radiations and vertebrate embryonic systems are, in the main, calculated to contribute to one or more of the following categories of study:

1. *Radiobiological mechanisms.*—To this category belong those investigations, the primary aim of which is the elucidation of processes stemming from the first impact of radiation on the developing system to the earliest and later chemical and biological responses; at this level the deliberations of the radiochemist become especially important to the radiobiologist. Practically all radiobiological studies on vertebrate developmental stages have something to contribute directly, or indirectly, to this category.

[1] The survey of literature pertaining to this review was concluded in February, 1956.

[2] The generous assistance of E. J. Frank, Phyllis Galasinski, and Barbara Herbes in many phases of the preparation of this review is gratefully acknowledged.

[3] It is a pleasure to acknowledge the kindness of Drs. Liane B. and W. L. Russell, and Dr. Samuel P. Hicks in permitting the reproduction here of some of their published and unpublished figures; Dr. Hicks and Dr. Robert Auerbach also generously supplied copies of manuscripts in press.

2. *Morphogenetic mechanisms.*—Here belong those studies whose primary aim is the clarification of the mechanisms and principles of vertebrate development. It is with this category that the present review will be chiefly, though not exclusively, concerned. Here also interests can be at the molecular level, but it will be clear that the viewpoint is not the same as in 1.

3. *Clinical embryology.*—This is neither a logical nor a natural category. It is rather a matter of convenience and will be so regarded. It includes immediate clinical observations and such implications for medicine as may be suggested from investigations more immediately related to categories 1, or 2, or both.

Many studies are concerned with more than one of the above categories; this need not militate against the usefulness of the classification. With the ever-growing volume of studies in all phases of radiation biology, it becomes increasingly important that individual studies be classifiable at least in terms of some general theoretical areas. The criticism is here submitted that in radiodevelopmental studies, as well as in various other phases of radiobiology and biology, authors fail with unwarranted frequency to point up clearly and unmistakably, in their introduction, the specific issue to which their experiment is directed.

An effort will be made in this review to discuss the more significant contributions of the past three years in terms of their pertinence to the categories defined above. That all publications cannot here be treated at deserved length is determined by limitation of space. Further, it would involve considerable duplication if this chapter were to be formally organized on the basis of the above three categories, desirable as such might be. As a matter of convenience, therefore, each vertebrate class will be considered separately. The major contributions deriving from studies within each class will, in turn, be channeled to the categories of study earlier defined. This, it would seem, is at once the least and the most that can be done in the limited space available. There is always the vexing problem of what will be excluded and which reports will receive either passing or preferred attention. The necessarily arbitrary solution to which a reviewer must arrive ought to be set down early. Accordingly, the scope of the present chapter is defined below.

Although intimately related to embryonic (radiodevelopmental) effects, genetic effects of ionizing radiations will not be considered. Here, the zygote at least is presupposed. Phenomena such as the "Hertwig effect," important though they be, will not be reviewed; for in this instance, the radiation is applied quite prior to and apart from the developing system, *per se.* By the same token, radiation effects on gametes (e.g., on spermatogenesis) or on the fertility of exposed adult animals will not be discussed. Further, and especially important, it was early decided that only studies involving irradiation from external sources would be included; there are cogent reasons for this limitation. Radioisotopes are being increasingly employed in the study of the effects of radiations on embryonic development at the molecular, cell,

and tissue levels; even a cursory examination of the literature will likely convince any reader that the study of the effects of radiations from internal sources has become so extensive and involved as to warrant its consideration in a separate review. This is not, in any way, to suggest that these methods are other than complementary in the problems at hand.

It is acknowledged that the above limitation of review coverage stands rather in contrast to Rugh's (1) earlier review in this same series, which included practically all types of radiobiological work which involved the use of vertebrate gametes, embryos or larvae; while we are extremely indebted to this excellent survey and commentary, we feel it might well have been more limited. L. B. Russell (2) has emphasized the importance of distinguishing, in radiobiological studies, between those investigations which are properly embryological and those which are not. An effort has been made to include all publications of the past three years which bear directly upon the subject of this review as herein defined. In principle, more properly developmental studies, and those reflecting more promising avenues of approach, have been accorded larger attention; it is hoped that no directly pertinent publications have been omitted.

This review covers studies published between February, 1953 and February, 1956. In addition to the many valuable individual research contributions appearing during this period, the following more comprehensive publications warrant special mention, for they embody important contributions highly pertinent to the present subject:

(*a*) *Symposium on Effects of Radiation and Other Deleterious Agents on Embryonic Development* (3) (sponsored by the Biology Division, Oak Ridge National Laboratory). This is a valuable collection of papers by distinguished investigators which has received high praise (4). The discussions of each paper have been included and constitute not the least stimulating part of the symposium.

(*b*) *Radiation Biology* (5) (edited by Hollaender). No praise can be too high for this distinguished successor to the pioneering *Biological Effects of Radiation* [(6) edited by Duggar].

(*c*) the excellent review on "Vertebrate Radiobiology: Embryology," prepared by Rugh (1) and appearing in volume three of the present review series. In the interests of historical continuity it may be noted that in his review Rugh did not mention the important older reviews on the effects of radiation on embryonic development and regeneration by Butler (7) and Curtis (8), respectively.

The several vertebrate classes do not enjoy equal distinction as experimental vehicles in radiodevelopmental research. In the literature, reptilian embryos are as conspicuous by their absence as are the mammalian embryos by their popularity; the unfortunate reptilian situation obtains in spite of the fact that their developmental stages are quite well known and the material is rather easily available. Equally regrettable is the only slightly less unpopular status of the fish embryo in such studies; Rugh (1) has suggested

some likely reasons for this. It is hoped that such deficiencies in the vertebrate (radiodevelopmental) series will be remedied in the future. In spite of the fact that avian development has been so well investigated, descriptively and analytically, this group (represented almost exclusively by the chick), has not in the past few years enjoyed the measure of currency in radiodevelopmental studies that might reasonably be expected. The amphibia (particularly anuran and urodele larvae) continue to maintain solid representation in this type of investigation. At present, however, mammalian prenatal stages unquestionably occupy a place of distinguished popularity in radiodevelopmental studies, particularly developing mice and rats. Undoubtedly the availability of genotypically well-recognized strains and, perhaps more importantly, increasingly improved methods for more accurate estimation of gestation stages in these small mammals, together with the quite understandable preference they enjoy by reason of the likely applicability of results and interpretations in this group to human embryological problems, have converged to establish the high popularity of this class in such investigations.

Fish

Only two relevant reports on fish embryos are at hand. Welander (10) has studied effects of 200 KVP x-rays on different embryonic stages of the trout, *Salmo gairdnerii;* his investigation was designed to ascertain the periods in development during which irradiation would be most damaging. The stages chosen for study were one-cell, thirty-two-cell, early germ ring, late germ ring, early eye and late eye; these represent development from 2 to 479 hours. Employing the LD_{50} technique, with 25 to 2570 r, it was found that the one-cell stage is 5 to 15 times more radiosensitive than any of the other five stages studied; Welander allows that the overall data are not easily interpreted and points out difficulties attending comparison of the results on the embryo of the trout with those of other fish and higher vertebrates.

Rugh & Clugston (11), employing 182 KVP x-rays, addressed themselves to the problem of determining the radiosensitivity characteristics of the eggs, sperm, early cleavage, and later embryonic stages of *Fundulus heteroclitus;* in this paper they also report some experiments involving point irradiation with "a silver point coated with polonium" allowing of alpha-irradiation of single cells in early cleavage stages and of localized areas of later embryos.[4] The authors found their results with x-rays to be in general agreement with the earlier work of Solberg (67), who pointed out that embryos of this species exhibit decreasing radiosensitivity with increasing age. The fertilized egg and early cleavage stages were substantially retarded by as little as 300 r; blastulae developed "fairly normally" after 1000 r while eight day embryos

[4] The authors indicate that this method of point irradiation with polonium was probably first used by Lacassagne and Desclin in 1928.

tolerated up to 3000 r without marked increase in the usual (control) percentage failing to develop normally. For details of their results following especially heavy irradiation (100,000 to 200,000 r) of eggs and sperm, and the possible relation of this work to Solberg's results, it is advisable to consult the paper. Point irradiation with polonium (20,000 rep) of a single blastomere of the two-cell stage appeared to damage the cell to such an extent that regulation and successful gastrulation could be achieved only in rare instances.

While both of the above studies have contributed valuable information, particularly in reference to the comparative radiosensitivity of different developmental stages (and gametes), it is likely that the results will not serve immediately or largely to improve substantially the position of the fish embryo as a vehicle in radiobiological studies. It is, however, to the credit of these investigators to have engaged this "unpopular" group. As yet, the "radiobiological niche" of the fish embryo has not been found, or at least not recognized.

Amphibia

As earlier indicated, developmental stages of the amphibia occupy a prominent position in the scale of vertebrate material subserving the analysis of radiobiological and morphogenetic mechanisms. In this section some attempt has been made to classify the studies considered in terms of certain areas of interest.

Regeneration.—Pestalozzi (12) studied the effects of 180 Kv x-rays, 1500–4000 r, on the regenerative capacity of the tail of larvae of Xenopus; total body irradiation was applied six hours after amputation. While the regenerates are smaller and the process proceeds more slowly after irradiation, "the regenerative power remains intact" even after 4000 r, and regenerative power persists until the death of the larva. Only striated muscle was found not to regenerate following irradiation. The point is made that while there are clearly regressive tissue changes in the regenerating portions following whole body irradiation, the larvae did not show any obvious damage in the "normal tissues" of the body. The author uses the term "normal" here quite unwarrantedly, for no such irradiated tissue can be rightly called normal; he would better refer to such as non-regenerating tissues. He allows that death which occurs within 9 to 18 days is probably due to the total irradiation combined with effects of trauma and the anesthetics employed.

Höhn (13) irradiated larval Xenopus exhibiting three, five and seven day old tail tip regenerates (3 mm of the tail was amputated) with 1550 r of 180 KV x-rays; she found that following irradiation all the larvae showed a quite normal regenerate. "Degeneration commenced on the 3rd to 5th day after irradiation and finally caused the death of the animal."

This reviewer does not clearly see that the above two studies on the effects of x-irradiation on vertebrate regeneration shed light on radiobiological or morphogenetic processes, significantly over and above what was, in gen-

eral, already known regarding the effect of larger and smaller doses of x-radiation on regenerative capacity.

Grossman (14) examined the effect of 500 r of 180 Kv x-rays on mitotic activity in the tissues of larvae of Xenopus (12 mm), at various time intervals following whole body irradiation. She found that mitosis was diminished two hours subsequent to irradiation and completely inhibited after 6 and 12 hours. After 24 hours it "recommenced" in all organs, even appreciably above normal in the gills. The general activity diminished again after 48 hours.

In connection with the general problem of cellular transformation during regeneration, Rose, Quastler & Rose (15) addressed themselves to the problem of determining whether the epidermis alone can furnish the cells for limb regeneration in urodeles. The authors review the earlier evidences relating to the problem and point up the necessity of a more rigorous test of the possibility of "epidermal transformation." Other workers have demonstrated that bone, muscle, and whole skin can each produce whole limbs; their experimental method consisted essentially in testing the developmental potency of a tissue by transplanting it to a limb which had been rendered incapable of regeneration by exposure to x-radiation. The present study involved an interesting variation of this technique. Both forelimbs of adult *Triturus viridescens* were irradiated below the elbow, with single doses of 500–10,000 r (150 Kv); the remainder of the animal was protected by lead. Left forelimbs served as controls; 7 or 28 days following irradiation these were amputated midway between the wrist and elbow. The right forelimbs were treated the same way except that at the time of amputation the whole skin was stripped off the remaining stump, up to the point of non-irradiated tissue 1 mm above the elbow. It was already known from previous studies that in such a stripped stump epidermis alone migrates distally and accumulates at the tip of the stump; accordingly, the epidermis that eventually migrated and covered the stumps of the right limbs in three experiments was non-irradiated. After a dose of 3500 r or higher, there was complete absence of regeneration on the control side, while the stumps on the right side, now covered with non-irradiated epidermis, showed some hand regeneration. The regenerates that occurred in the right limbs must have come from the non-irradiated epidermis. The authors write as follows:

> It is concluded that epidermis can serve as the only source of cells for a regenerating limb. It now appears likely that any limb tissue can serve as a source of regeneration cells. Whether one is the predominant source both in larvae and adults is not clear . . . .

This study is an illustration of the profitable use of x-rays as a tool in the elucidation of morphogenetic processes. The paper, brief though it may be, is also a good example of clearly defining a specific issue at the start and focusing upon the same to the end.

Rieck (16) studied the effects of single doses of ultraviolet on the fore-

limb of larval Amblystoma in Harrison stages 29 to 43. It was demonstrated that there are definite and pronounced effects of localized irradiation on the developing limb bud; the sum total of abnormalities increases as the degree of differentiation in the developing limb, at the time of irradiation, increases. That is to say, susceptibility to ultraviolet increases with progressing differentiation. This is extremely interesting and stands in remarkable contrast to earlier studies with x-radiation wherein it was found that x-ray inhibition of limb development decreases with progressing differentiation of the limb primordium. The implications of this aspect of Rieck's study can only be gathered by reading the text. It is especially interesting that the results here show that retardation of distal limb structures does not greatly interfere with differentiation of proximal parts, as has been demonstrated by others with entirely different techniques, for the amphibian and avian limb. The data on photorecovery in this study are discussed below.

Butler & Blum (17) have recently investigated an important aspect of the effects of ultraviolet on regenerative growth in urodele larvae (Amblystoma). Their study was specifically directed to the development of accessory limb structures from blastemata formed subsequent to localized exposure of the larval limb to ultraviolet; studies on limb regression and photorecovery were also involved (see their paper for details regarding dosage). Photorecovery is discussed in a section below. The authors reserve a detailed description of the anatomical characteristics of accessory limb structures to a later communication; their data on post-irradiation formation of blastemata will be our principal concern here. When the complete elbow region is irradiated there ensues, in the irradiated region, a process of tissue regression with consequent shortening of that part of the limb (forearm) involved; this shortening may be accomplished at 3 to 4 weeks following exposure. The authors observed that

> During the fourth and fifth weeks a new type activity begins in the irradiated region. It is during this period that a protrusion may appear near the elbow or along the posterior border of the forearm. In its manner of development and in the potentialities which it later manifests, this protrusion resembles a regeneration blastema in every way . . . Once a blastema is established it grows rapidly in size and soon begins to exhibit morphogenetic capacities. Differentiation takes place within the blastema and one or more digits appear.

The incidence and morphological characteristics of accessory structures vary according to site of the localized exposure. This is a stimulating study and undoubtedly a very promising attack on the important problem (among others) of the relationship between the short lived process of regression and subsequent regeneration; the further outcome of these investigations will be awaited with interest. The relation of studies such as the above to problems of neoplastic growth is not difficult to see. It is highly desirable that the employment of ultraviolet in studies on morphogenetic mechanisms would enjoy increasing currency. This type of radiation has, in many situations,

a distinct advantage over x-rays; it frequently provides a more delicate and wieldly experimental tool.

Growth and miscellaneous.—Fritz-Niggli (18) investigated the "biological depth effect" of 180 Kv x-rays as compared with ultrahard radiation produced by a betatron at 31 Mev, on the development of larval Xenopus. He found that 2000 r at 31 Mev, "measured and given at the maximum intensity of 5 cm," affected the development of the larvae up to a depth of 30 cm in a water phantom, whereas the 180 KV radiation, "2000 r at the maximum intensity at the surface," showed no effect beyond a depth of 5 cm. This is interpreted as confirming the physically measured "great depth effect of the ultrahard betatron irradiation"; there would appear to be no reason for taking any exception to this interpretation.

Piatt & Raventos (19) have contributed a study on the effect of transplantation of certain structures from irradiated urodele embryos to non-irradiated host embryos of the same species. Embryos of *Amblystoma punctatum*, stages 15 and 16, were exposed to 200, 400 or 600 r of 200 Kv x-rays. Four days after irradiation (stage 27), three series of orthotopic transplantations were made, employing the irradiated embryos as donors and non-irradiated embryos of same stage as host; the otic vesicle, medulla without ear, and forelimb were the transplanted structures. Development of the transplanted irradiated ear grafts "ranged from fairly normal to complete absence"; in general, however, the transplants attained a much higher level of development than was achieved by these same structures when they were left in totally irradiated animals. The medulla grafts exhibited something of the same pattern of response when compared with medullae not removed from totally irradiated embryos. The situation with transplanted forelimbs is (to this reviewer), a bit confusing. The authors interpret the results of transplanting these irradiated tissues to a normal developmental environment as indicating "that the intrinsic potentialities for post-irradiation growth and differentiation cannot be assessed properly by observing the fate of the part in the unhealthy environment created by total body radiation."

A few comments on the above paper are indicated. Apart from its scientific merits, its "introduction" is an example of lack of attention to the matter of pointing up the specific issue to which the investigation was directed; historical details are plentiful but there is a well nigh complete lack of clearly stated questions to which the reader can relate subsequent details. The questions are there "between the lines," but it would be better if they were in the lines. In the introduction the authors review briefly the work of Schneller (20) on chorio-allantoic grafts of x-irradiated chick primordia; in substance, her findings in this regard were as follows: within three hours following irradiation she excised organs from chick embryos previously exposed to a lethal dose of x-radiation, and transplanted these to the chorio-allantoic membrane of normal host chick embryos; irradiated donor optic vesicles and limb buds (from early embryos) survived and developed in a

manner comparable to control grafts. Among other things, this suggests that these organs themselves can tolerate a dose which is lethal to the embryo as a whole. Piatt & Raventos indicate that Schneller suggests that x-rays produce their effect on the organism functioning as a unit; they then add: "If this be true a correlative inference would be that the indirect effect of irradiation is more important than the direct effect." This, as it is stated, would seem to be a somewhat unwarranted inference and would likely be so regarded by at least some others familiar with Schneller's work, and familiar also with frequently poorly oriented arguments regarding the relative importance of "direct" and "indirect" effects of radiation in a particular syndrome. It will be clear that in this type of investigation time is a most important element. Although Schneller made her transplants within three hours after irradiation and these workers made theirs only four days after exposure, it is stated by the latter that, in general, their experimental approach and procedure were similar to Schneller's, although their results were somewhat at variance; later in the paper they allow that this time factor constitutes a "significant difference" in the respective procedures. Considering the nature of the problems involved, it is difficult to agree that these procedures and approaches are methodologically similar. Other things being equal, this particular time-interval is, it would seem, an extremely important factor; the differences in the results of these two investigations is in no way surprising. Piatt & Raventos submit that this time-interval difference "may account for the difference in our results"; the present reviewer would enlist this as a solidly probable explanation. Finally, it is very difficult to agree with these authors' insistence that their data "lend no support to the concept of indirect radiation effect upon the development of early embryonic tissues." On the contrary, it is here submitted that their data, viewed with those of Schneller, provide excellent evidence for such indirect effects.

Apart from the above criticisms, Piatt & Raventos have made a valuable contribution. Like Schneller's work their investigation serves to emphasize, among other things, the fact that when organismal death issues from a radiation insult, the result is likely due in largest measure to the hyper-radiosensitivity of one or a few organs or organ systems; other organs and systems, though in themselves actually quite radio-resistant, cannot compensate for the overall effect of radiation on the hypersensitive ones, particularly such as the vascular system. From the viewpoint of both radiobiological and morphogenetic mechanisms, this is an extremely promising avenue of approach.

Brunst (21) has studied two aspects of the effects of localized 55 KVP x-rays on the anterior part of the head of the young axolotl Siredon; namely, the effect of radiation on the eyes and the effect of such localized head irradiation on the growth of the animal as a whole. He found, with 4000–8000 r, that animals of different ages exhibited, after irradiation of the anterior part of the head, a local suppression of growth in the exposed region

and an inhibition of growth of the animal as a whole. Both of these observations are in accord with results in earlier studies on small mammals. As might reasonably have been predicted on the basis of previous studies, the cells of the retina were found to be the least radioresistant, whereas the pigment cells of the choroid were remarkably radioresistant.

A few months later Brunst (22) published another paper, of approximately the same length, on the influence of x-rays on the development of the eye of the axolotl, in which paper no reference is made to his immediately preceding work on this subject (21). It appears that the two papers were intended to be substantially the same, for the observations and discussions are quite similar and several photomicrographs are identical. In the later paper, however, there is added a brief discussion on the significance of the data for practical roentgenology.

The problem of the reaction of skin epithelium of the tail or larval *Rana catesbiana* to 2000 and 4000 r of locally applied 60 Kv x-rays was investigated by Brunst (23). In one group the radiation was applied to the terminal region following development of a 16 to 18 day old regenerate; in a second group the tail was similarly irradiated but no amputation was involved. The formation of "degenerating giant cell epithelium" was observed in both groups; however, this was seen in approximately 2½ times as many cases in that group which had undergone amputation and somewhat extended regeneration prior to irradiation. Characteristics of the typical giant cell epithelium are described in some detail. The formation of unusually large outgrowths of epithelium, consisting of cells of normal size with numerous mitoses, were sometimes observed between portions of degenerating giant cell epithelium; the author offers this as an example of "indirect roentgen stimulation."

A comparative study of the effects of 2000 to 4000 r 55 Kv x-rays on jaw and tooth development in young axolotls (20 to 60 days) and mice (1 to 3 days) was engaged by Brunst (24). It was found that the biological response to localized irradiation of the jaws in both forms issued in much the same effect; in short, both the growth of the jaw and the development of teeth was inhibited. The author states:

> The most remarkable outcome of these experiments, however, was that the teeth and partially developed teeth present at the time of irradiation disappeared. Roentgen irradiation of the jaws thus destroyed or inhibited not only the mechanism of tooth formation but also the mechanisms of tooth maintenance.

(We may assume that the employment of the singular and plural of "mechanism" above were not intended to suggest any likely simplicity of the developmental process as compared with the maintenance process.) A bit further it is observed: "There is thus a striking similarity in the biological reaction of irradiated jaws in two such widely separated forms as mice and axolotls." It is not altogether clear why this similarity should be regarded by the author as "striking," since, in his introductory remarks, he allows

that " . . . the process of tooth formation is essentially the same in most vertebrates . . . " To this reviewer it would be remarkable if, with the dosages employed, the teeth and partially developed teeth had not eventually disappeared. Furthermore, the overall similarity in the response of representatives of these two vertebrate classes, to the experimental conditions defined, is hardly surprising; it would, we suggest, be a remarkable and striking thing if the respective responses had been significantly different. Nevertheless, it is informative to know that this particular similarity in response has been experimentally demonstrated.

Radiosensitivity.—Rugh (25) has made a valuable contribution to the radiodevelopmental study of amphibian embryos. The first half of this paper consists of a quick review of radioembryological studies on amphibia that are included in his original review in this series (1). We wish to call attention to his more extended discussion of Brunst's (26) concept of "stimulation of mitotic activity by roentgen irradiation," which, Rugh submits, "cannot yet be accepted as proven." The second part of Rugh's paper describes his own results on a number of problems relating to the radioembryology of the frog (presumably, *Rana pipiens*). He used 110 KVP x-rays. Some of his findings are as follows:

(*a*) Frog tadpoles can be killed within one and one-half hours by 160,000 r or more. Suspecting that some clue to the causes of this acute radiation death might be reflected histochemically, he found that immediately following irradiation, irradiated tadpoles exhibited a drastic depletion in their reserves of mucopolysaccharides, particularly in the periphery of the central nervous system, the eyes and muscles.

(*b*) Cysteinamine (dissolved in the swim water) prolongs for a short time the life of tadpoles exposed to a lethal dose of x-rays; however, it does not protect them against the lethal effects of exposure. Rugh indicates that the results in this connection were "inconclusive but suggestive"; they indicate that this type of investigation into chemical protection against radiation effects on amphibian developmental stages ought to be extended.

(*c*) The various developmental stages of the frog, in order of decreasing radiosensitivity, can be listed as follows: 1. fertilized egg; 2. blastula; 3. first cleavage; 4. uterine egg; 5. gastrula; 6. yolk plug; 7. the ovarian egg; 8. the adult frog; 9. the neurula; 10. stage-23 tadpole; 11. premetamorphic tadpole. The determination of these comparative radiosensitivities occupies the larger part of this study and while the data are submitted as essentially qualitative, quantitative inferences can be made.

(*d*) On the basis of what is acknowledged as circumstantial evidence, it is concluded that the differentiating cell, even prior to mitosis, is more radiosensitive than the differentiated cell actually in the process of mitosis; the need of more direct proof is pointed out.

(*e*) A latent period always obtains between exposure to radiation and the manifestation of any developmental abnormality; pycnosis or karyorrhexis, however, may be observed immediately following exposure.

(*f*) The most frequent types of abnormalities observed in irradiated embryos are: pycnosis or karyorrhexis of differentiating cells, exogastrulation, edema, flexure, aneurogenic development and amorphous development.

Rugh concludes with the following:

> It can be said that any embryonic stage (excepting the tadpole) is infinitely more radiosensitive than the adult of the same species, by any criterion of estimation. This may be related to the protracted period of differentiation of the central nervous system and its accessories.

Voino-Yasenetskii (27) investigated the effects of gamma radiation, from cobalt-60, on ovulation, fertilization, and embryonic development of the frog. Mature male and female frogs were exposed (in winter) over a period of 48 hours to a sublethal dose of between 6800 and 7500 r. Irradiated and non-irradiated females received abdominal injections, 2 and 12 days after irradiation, of suspensions of hypophyses taken from both irradiated and non-irradiated females; following all experimental situations the speed of the ovulation process and the quantity of eggs ovulated were identical with these same manifestations in control animals. Artificial fertilization was effected in a variety of combinations of gametes from irradiated or non-irradiated animals, following ovulation induced with suspensions of irradiated or non-irradiated hypophyses. It is impossible to summarize the experimental details here; after considering all the data, however, the author submits that, as with the process of ovulation, the fertilization process was not disrupted by ionizing radiation, with the dosage employed. However, other conditions being equal, embryogenesis was more "strongly disrupted" when ovulation had been induced by hypophyses taken from irradiated females. The author summarizes:

> It can be concluded from this, that, under normal conditions, the function of the hypophysis has an effect not only on the quantitative aspects of the process of ovulation, i.e., the speed of the process and the number of ovulated ova, but also on its qualitative aspects. The quantitative aspects, with the doses of irradiation used in these experiments, are not disturbed, but the qualitative aspects suffer severely. Therefore, it must be recognized that one of the mechanisms which disrupts embryonal development, and which occurs even outside of the maternal organism, is damage by penetrating irradiation of the function of the maternal hypophysis.

In a continuation of their important studies on factors influencing the radiosensitivity of hematopoietic cells (in the kidney) of the frog tadpole,[5] Allen, Schjeide & Piccirillo (28) report briefly on the influence of subsequent anoxia on such cells previously exposed to x-rays or colchicine. The anoxia was produced by placing the larvae in a shallow, covered plastic box filled to overflowing with water which had been boiled for 30 minutes. Anoxia

[5] Tadpoles of *Rana catesbiana* are indicated as the experimental material, but in the legend to the graphical representation of results a species of *Bufo* appears to be given the credit.

reduced the percentage of cell breakdown following x-irradiation and equally reduced the mitotic arrests in tadpoles previously receiving tail injections of colchicine. In short, the authors show that anoxia also suppresses cell division and thereby retards x-irradiation damage; the period of suppression of cell division occurs roughly in proportion to the degree of anoxia induced.

Ultraviolet and photorecovery.[6]—Since its recent discovery only a very few investigations of this matter of photoreactivation have involved the use of developmental stages of vertebrates. It would seem that it is a phenomenon which is eminently calculated to contribute much to our knowledge of radiobiological and morphogenetic mechanisms.

To Blum and Matthews (30) goes the credit for first demonstrating this phenomenon in a vertebrate. (This contribution was not noted in Rugh's earlier review (1) in this series.) These authors found that illumination with wave lengths between 0.3 and 0.5μ greatly increased both survival time and recovery of capacity for growth and regeneration of larval Amblystoma which had previously been subjected to repeated small doses of ultraviolet radiation; this recovery occurred only when photoreactivating light was applied after ultraviolet irradiation. Of considerable additional interest is the fact that recovery could be demonstrated when the photoreactivating light was first applied as long as 20 hours following exposure to the ultraviolet; in this case, however, the degree of recovery was less than that issuing when the illumination was applied earlier. The authors also discuss briefly the evolutionary implications of the phenomenon of photorecovery in living systems.

Some two years later Rieck (31), in an investigation embracing considerably more than photoreactivation studies, demonstrated photorecovery from certain deleterious effects of ultraviolet irradiation (single exposure) on limb buds of Amblystoma; all the damaging effects of the ultraviolet were not (always) counteracted by visible light. This demonstration of photorecovery in the vertebrate limb bud also provided evidence that the depth of penetration of the ultraviolet radiation involved was greater than that assumed by some earlier investigators.

More recently, Butler & Blum (32) studied photorecovery following localized ultraviolet irradiation of areas of the limb of larval Amblystoma. They found that illumination with light of known photoreactivating quality, following ultraviolet irradiation of the elbow region, was accompanied by a marked decrease in the extent of tissue regression and consequent shortening of the limb which characteristically occur in an irradiated region. They also observed that survival of larvae kept in light, following probable exposure of

[6] Photorecovery (or photoreactivation) is the term applied to the phenomenon discovered by Kelner in 1949 "in which changes produced in different types of organisms, mainly microorganisms, by ultraviolet radiation of wave lengths around 2500 A can be counteracted if the irradiated organisms, under proper conditions, are exposed to a radiation of longer wave length, in the range between 3300 and 4800 A" [Dulbecco (29)].

considerably more than the limb region, by reason of technical difficulties, was much greater than similarly exposed larvae which were subsequently kept in the dark; larvae not subjected to ultraviolet survive equally well in the dark or light. These latter observations support the earlier conclusions of Blum & Matthews (30). While their analysis is not yet complete, Butler & Blum, in this study, have good evidence to show that the development of accessory limb structures is much reduced when animals are situated in light for a few days following ultraviolet irradiation. We concur with their estimate that it seems evident that ultraviolet irradiation combined with photorecovery provides a method for the study of numerous problems of developmental growth. As earlier suggested, it also provides a valuable tool for the elucidation of radiobiological mechanisms.

While the study is not directly related to radiodevelopmental problems, it may be noted that Rieck & Carlson (33) have recently demonstrated photorecovery in the albino mouse, thereby extending the observation of photoreactivation phenomena to a second class of vertebrates.

Birds

Avian embryos are represented here in disappointingly limited measure; as might be anticipated, the chick embryo bears the whole burden.

In view of the desirability of some standardization of methods for measuring the LD_{50} of chick embryos, as at least one indication of the biological efficiency of a given type of radiation at a particular dose-rate, Boland (34) studied the lethal effects of x-rays on chick embryos, particularly from the viewpoint of the mode and time of death. He investigated the bearing of these manifestations on the determination of the LD_{50} of the four-day chick embryo and found 4 time intervals, subsequent to exposure to different doses, during which it is particularly disposed to die; death following each defined interval was regularly associated with a "characteristic mechanism," reflected in specific patterns of hemorrhage. With 250 Kv x-rays at 155 r/min., the LD_{50} for four-day embryos was found to be 887 r for death within six hours following exposure and 717 r for death within ten days. This study was well controlled and is an important contribution to the methodological problem of the measurement of LD_{50}s of the chick embryo.

Schneider & Cheever (35) investigated the growth of vaccinia virus in tissue cultures of x-irradiated skin, muscle and cartilage of 6 to 14 day old chick embryos, with a view to determining whether radiation effects in the host cell (viruses being obligate intracellular parasites) might be reflected by changes in the amount of viral activity in the fluid or tissue or both phases of the cultures; they found that irradiation of such tissues *in vitro* had no significant effect on their ability to support viral growth, whereas irradiation of the tissues *in ovo* "appeared" to diminish their ability to support growth of the virus. They regard their data as suggesting that the differences accompanying *in ovo* and in *in vitro* irradiation were "due chiefly to the secondary effects of radiation rather than to primary ones." In an-

other place in this paper the authors refer to these effects as "secondary radiation effects."[7]

Goldman, Glasser & Tuttle (36) directed their attention to a study of the effects of radiation on the extraembryonic circulation of chick embryos; they used 600 to 800 r of 250 KVP x-rays. Injury to vascular components could be detected within a few minutes following irradiation. At 20 hours subsequent to irradiation there was a 62 per cent mortality of the embryos; beyond 48 hours none survived. Injury to capillaries was most conspicuous. Extraembryonic vascular deterioration was accompanied by intraembryonic hemorrhage. The prominent feature of their observations was the disintegration of extraembryonic vessels prior to the appearance of frank blood in that region. This study appears to be in its beginning stages; the authors are currently investigating biochemical mechanisms conceivably operative in these vascular deteriorations. Studies calculated to enhance our knowledge of the effects of radiation on vascular systems must always be regarded seriously; it would seem that enlistment of the easily observable and readily accessible extraembryonic vessels of the chick offers many possibilities.

In Bacq & Alexander (37) there is reported a very interesting case of chemical protection from x-ray damage in bird embryos. On page 312 they write:

> Wolff has found what may prove to be an example of direct cellular protection (i.e. similar to that found with unicellular organisms) in the case of birds. After exposing specific parts of bird embryos to x-rays, monsters (e.g., cyclopis, anophthalmia) are regularly produced during the subsequent development. If the embryo has been previously injected with cysteamine the number of monsters which appears is greatly reduced.

The source of Bacq & Alexander's information is a personal communication from E. Wolff. We have found no reference to publication of this work; it is likely that the material was the chick embryo. More work of this type ought to be undertaken. [See Rugh & Clugston (45) on cysteinamine protection of fetal mice.]

Mammals

Auerbach (38) exposed pregnant female mice to 300 r of 250 KVP x-rays nine and one-half days after the appearance of a copulation plug; embryos were fixed at half-day intervals following irradiation. A variety of abnormalities were recorded but the author has concentrated on spina bifida. He writes: "It is interesting to compare the x-ray results with hereditary spina bifida in the mouse, in an attempt to understand more clearly the nature of this abnormality. Spina bifida occurs in response to several muta-

[7] In the opinion of this reviewer, these authors do something of a disservice to their contribution by a rather confusing use of these terms. Perhaps for their "secondary radiation effects" the term "general indirect effects" might be less confusing; after all, "secondary radiation" is a respectable physical term and certainly the authors are not speaking of the consequences of this.

tions with strikingly different initial developmental disturbances; a common factor of these genetically determined types of spina bifida, however, may lie in an interference in the developing mouse embryo with normal formation or maintenance of neural tissues of the lumbo-sacral region. The x-ray results suggest that such interference at an appropriate time in development may in itself be sufficient to result in abnormal dependent differentiation leading to spina bifida."

The effects of single and fractionated doses of x-rays on mouse embryos has also been investigated by Auerbach (39). Pregnant females, nine and one-half days after appearance of a copulation plug, were exposed to either (*a*) a single 300 r dose or (*b*) three 100 r fractions at intervals of 30 minutes (250 KVP). Embryos were obtained by dissection 3–6 days following irradiation; the criteria of damage were lumbosacral spina bifida and coloboma of the eye since these occur with high frequency following irradiation on day nine and one-half and are easily classifiable. The incidence of these malformations is not affected by fractionation of the dose as employed but the severity of the defects is far greater following a fractionated dose than in response to a single dose. This interesting observation is discussed briefly in its possible relation to radiological practice.

With mouse fetuses of the same age as in the two studies above, Auerbach (40) has made a particular study of coloboma of the eye. Following 300 r of 250 KVP x-rays, coloboma of the eye was observed in 85 per cent of the embryos; histological damage was detected within one and one-half hours after irradiation. Further development of coloboma is similar to hereditary coloboma. This work will be published soon.

Brunst & Figge (41) investigated the effects of localized irradiation of young mice. Employing 3000 and 4000 r of 55 Kv x-rays and mice of 1 to 25 days of age (mostly 1 to 4 day old animals), they irradiated the head alone in one group and in a second group a narrow transverse band midway between fore and hind limbs. In the first group, as early as a week following exposure, suppression of both head development and development of the body as a whole was observed; disturbance of brain function was reflected in "paroxysms" occurring nine days after irradiation. In the second group paralysis was seen in some of the 1 to 4 day old mice so irradiated and then usually in those individuals which had been irradiated "exactly in the region of the motor neurons controlling the hind limbs." In the first group it was difficult to find histologic evidence of damage to the brain; in the second group, upon histologic examination, the irradiated spinal cord region in most cases appeared normal, while in others it is completely destroyed.

Rugh & Wolff have studied some responses of the fetal eye of the mouse to various doses of 210 KVP x-rays (42, 43). These two studies are quite similar. In one, the 13½ day fetus was used and the eye examined histologically and cytologically at 4, 24, and 72 hours and 6 days following exposure of pregnant females to 300 r; in the other the 12½ day fetus was studied following doses ranging from 50 to 300 r and at intervals the same as above,

with a six week interval added. It seems convenient here to consider these studies as a unit. The most important observation emerging from this investigation is the remarkable reparative power of the fetal eye following severe radiation insult, in spite of the fact that the developing eye is notoriously radiosensitive.

The presumptive retina can be seen to have sustained serious damage as early as four hours following exposure to 250–300 r; at 24 hours the damage is still considerable but some of the cellular debris has been disposed of by phagocytosis; by 72 hours the developing retina appears to be quite normal and at birth (6 to 7 days after irradiation) there is no gross evidence of radiation damage. An exposure of 50 r was without discernible effect on the eye. The more severe the radiation insult the earlier is there evidence of "cytological radionecrosis." The authors rightly emphasize that the data here are not evidence of recovery in the sense that the damaged elements regain their "integrity and activity." Among other things, this interesting study points up the importance of studying responses over a series of time intervals; if, for example, this study had been confined only to observations four hours after irradiation, or only to the 72 hour post-exposure interval, one might easily have been led to some rather erroneous conclusions.

The effect of a single sub-acute x-ray exposure (300 r; 210 Kv) on the subsequent skeletal growth of the 15½ day mouse fetus was the subject of a unique quantitative study by Levy, *et al.* (44). Skeletal relations, based upon the growth of three major bones of different embryological origin—femur, mandible and parietal—were analyzed statistically in both normal and irradiated animals. Experimental and control litters were sacrificed at regular intervals ranging from 1 to 240 days post partum. While the irradiated animals were born with the three bones in question exhibiting smaller dimensions than in normal animals, the proportionality of their dimensions was not affected by the radiation and, ultimately (eight months post partum), they each attain almost normal size. We quite agree with the authors in their persuasion that this study supports the thesis, suggested by earlier works of Russell, Wilson & Hicks, which they cite, that in the 15 day mouse fetus the skeletal bed is less radiosensitive than it is earlier, and that the radiobiological effect at this time is on growth (accretion) rather than on differentiation and morphogenesis. While there have been many excellent studies in the recent past on skeletal effects following irradiation of different developmental stages of rats and mice[see Russell (50)] this present investigation appears to have been the first to employ this type of quantitative approach. Data from studies such as this are invaluable in the interpretation of certain clinical facts, particularly such as are being carried out by the Atomic Bomb Casualty Commission in Japan.

In view of the rapidly rising interest in chemical protection against ionizing radiation, it is a pleasure to note what is probably the first published study relating to such protection in the fetus. Rugh & Clugston (45) wished to determine whether the protection known to be afforded the

irradiated adult mouse by previous intraperitoneal injection of cysteinamine (beta-mercaptoethylamine) could be transferred to the fetus. Female mice, on gestation days 13½ to 19½ were exposed to 300 to 1200 r of 210 KVP x-rays; cysteinamine was administered intraperitoneally to the pregnant mice 20 minutes before irradiation. It was found from a study of the data on both survival and weight at one month that this —SH compound provided some protection against x-rays in the fetal as well as the adult mouse. The authors suggest that related —SH compounds might lead to similar results in the matter of fetal protection. This is a stimulating study and it may be hoped that it marks the start of extensive studies on chemical protection of mammalian developmental stages against radiation damage.

In the past three years, two papers have appeared on the effects of x-radiation on embryonic development in the rat by Wilson, Jordan & Brent (46, 47); these were in press at the time of the preparation of Rugh's (1) review but were briefly considered therein. A third paper by Wilson (48), part of the ORNL Symposium, was largely based on the two previously noted publications [and also on Wilson & Karr (66)]; it also includes some observations not published up to that time. It therefore seems both convenient and reasonable, here, to cover the above publications by a consideration of Wilson's (48) most recent paper. The observations reported concern the effect of x-irradiation (12.5 to 600 r; 85 KVP) of rat embryos at days 8, 9, 10, and 11 of gestation. [Wilson & Karr (66) is best consulted for details of the experimental method.] The important point to be noted here in reference to method is that, following careful estimation of gestation ages, one uterine horn was exteriorized (by abdominal incision of the mother) and only a portion of the total implantation sites (embryos) irradiated; the other uterine horn (control) was similarly manipulated but without irradiation. Irradiated and control embryos were removed to one several days later, or at term; these were compared in reference to weight, malformations and rate of intrauterine mortality. Irradiation on day 8 had a qualitatively different effect on subsequent embryogenesis than issued from similar treatment on days 9, 10, or 11. After doses less than the totally lethal one (200 r) on day 8 the only "residual effect" was retardation of growth. Exposure at the older stages (day 9, 10, and 11) resulted in an increased number of malformations, increased mortality rate and retardation of growth as the dosage was increased. The difference in the pattern of response is attributed to a change in the radiosusceptibility of the cells of the embryo as it moves from an undifferentiated stage on the eighth day to the early stages of differentiation which begin on the ninth day. It is important to note that on day 8 the embryo consists "merely of a randomly arranged mass of blastomeres at the embryonic pole of the egg cylinder." By day 9 the primary germ layers have been laid down, the first large step in differentiation. The quantitative differences are attributed to " . . . an increasing threshold of sensitivity to the same radiation-induced effects as differentiation proceeds, once it has started." This interpretation must

not be taken to imply more than is intended. Part of the author's comments on the results will serve to clarify the point; he writes:

> The manner in which the susceptibility to irradiation was influenced by the degree of differentiation is unknown. It has been often generalized that susceptibility to irradiation tends to be inversely related to the degree of differentiation. The decline in sensitivity observed as differentiation progressed from the ninth to the eleventh day in the present experiment is in accordance with this generalization. An exception to the rule, however, is the more limited response of the eighth day as compared with the ninth–, tenth–, and eleventh-day embryos. It appears, therefore, that the diversity of reaction cannot be explained simply in terms of greater or lesser degrees of differentiation.

Although Warkany's paper (49) at the ORNL Symposium was primarily concerned with such disturbances in embryonic development of rats as accompany maternal vitamin deficiencies, we should like to point out that in this paper he discusses some earlier collateral experiments from his laboratory in which x-rays were used as the teratogenic agent. He points out, for example, that while certain malformations were common to both x-ray and riboflavin-deficiency induced syndromes, the overall variety of anomalies is greater in the x-ray syndrome.

Since early 1953, L. B. & W. L. Russell have authored four publications which are singularly important in this field of radiation effects on mammalian embryonic development.

We consider first L. B. Russell's review (50) entitled: The Effects of Radiation on Mammalian Prenatal Development. In general it may be said that a study of these 57 pages would alone give one a more than respectable introduction to this field. It is the most complete coverage of the literature available; the most that can be done here is to give some idea of its content and construction. There are five major headings: I *Introduction.* Here, in addition to pointing up the importance of this problem, the difficulties peculiar to mammalian embryos as experimental material are outlined. II *Experimental findings.* For reasons explained by the author these are considered under three headings, based on three broad phases of the general problem: (A) irradiation during the preimplantation period; (B) irradiation during the period of major organogenesis, and (C) irradiation during the fetal period—the period of growth and minor organogenesis. III *Mechanisms of radiation effects on the mammalian embryo and fetus* [more extensively and systematically discussed in Russell & Russell (51)]; here the following items are reviewed: (A) influence of the maternal organism; (B) nature of the primary damage and intermediate effects; (C) dosage relations; variability; (D) comparison with other agents affecting development. IV *Clinical literature on the effects of radiation on embryo and fetus; human implications of experimental work.* And finally the summary and, of course, a bibliography. (Some of the references go back almost 50 years.) In closing the author sets down fifteen summary statements which, taken together, constitute as fine a digest statement of the status of this field as

one could reasonably expect; it covers slightly more than two pages. No attempt will be made here to summarize further; the chapter is essential reading for all who are interested in this subject.

Next to be considered is the paper of Russell & Russell (51) entitled: An Analysis of the Changing Radiation Response of the Developing Mouse Embryo, which paper is part of the ORNL Symposium and is based largely on their own work dating from 1949. Here this skilled team (embryology and genetics) make a valuable contribution toward bringing into focus a general picture or pattern derivable from the large amount of information available; also included are some speculations on mechanisms by which radiobiological effects are realized. Overall, their studies contribute principally to two categories of interest: (*a*) the mechanism of radiation effects on developing mammalian systems and (*b*) experimental analysis of mammalian embryology, wherein the radiation is used merely as a tool. The larger part of their deliberations in this paper deal with the former issue. One of the most important points considered is the concept of "critical periods"; on this point the authors state (in part):

> Before advancing any hypotheses of basic radiation action, it should be made quite clear that use of the term "critical periods" does not imply that the immediate primordium of the character malformed as a result of irradiation at a certain stage was damaged at that stage, i.e., during the critical period. There are many cases of abnormalities that are obviously secondary to other abnormalities (e.g., sternal malformations, which are probably due to faulty growth of ribs, which, in turn, results from vertebral defects). There are probably even more cases in which the final malformations are the results of more obscure and as yet unsuspected chains of reactions. What a "critical period" does indicate is that the initial disturbance that touches off a chain of processes, regardless of their complexity, must occur at a certain stage to give a certain end result.

Viewing the data in an overall way, it seems clear to this reviewer that the concept of "critical periods", as characterized by these authors, is based on strong evidence (i.e., on their own data and that of others). This "critical period" problem will likely and understandably be the subject of considerable study in the future. While more extensive graphical representation of critical periods for the induction of various abnormalities appear in Russell (50) we present here in Figure 1 a typical example of such analyses, referring particularly to skeletal abnormalities.

In reference to the question of the mechanism of radiation effects on embryos the authors write:

> From the results of varied work on other organisms it may be concluded that the biologically effective damage of the generally disturbed ionizations is probably in the nucleus of cells since doses of considerably greater magnitude than those employed in this experiment are required to cause detectable cytoplasmic changes . . . . The most plausible interpretation is that the primary intracellular effect of radiation on the embryo is the induction of somatic chromosomal aberrations which result either in a breakdown of the mitotic process or in aneuploidy after cell division, the

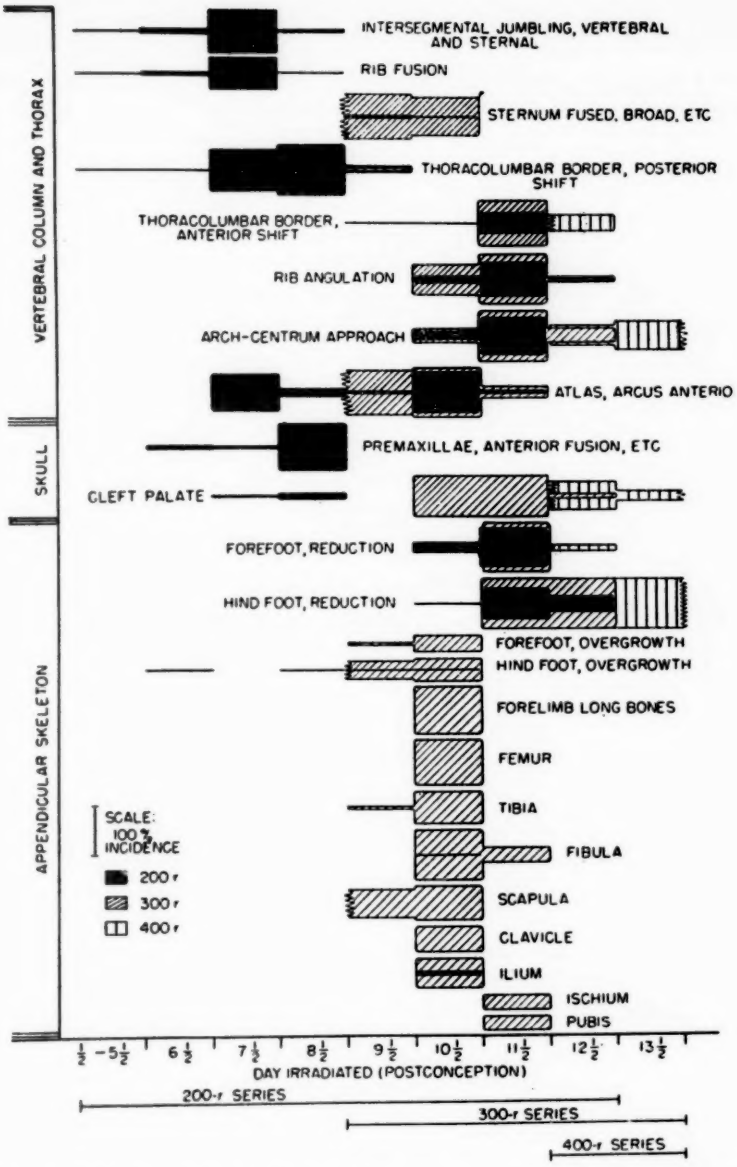

FIG. 1. Critical periods for the induction of a few of the well over 100 types of skeletal abnormality observed at term following prenatal irradiation. Representation is by (*a*) percentage incidence of abnormality (see scale in Fig.), and (*b*) magnitude of dose required to produce abnormality. Thus, the wider and more heavily shaded a band, the greater the sensitivity. Absence of a band at a particular stage indicates that the abnormality did not occur in the irradiated group, except where the serrated end of a band indicates that the dose series was not continued to the stage in question (see also bottom of figure showing extent of different dose series) [Courtesy Russell & Russell (51)].

end result in both cases being a cell lethal action. This explanation reconciles the observed consistency of response with the randomness of intracellular damage which must be postulated.

And they summarize their thinking on possible pathways in the development of malformations following irradiation of developing mammals in Figure 2; they intend it to emphasize the importance of caution in deducing the initial damage from the malformation observed.

Limitations of space preclude our giving here an even minimally respectable summarization of this paper and the interesting points brought up in its subsequent discussion by other participants in the symposium. We should like to add that this paper is a good example of clearly pointing up issues at the start and orienting the whole presentation to these defined questions.

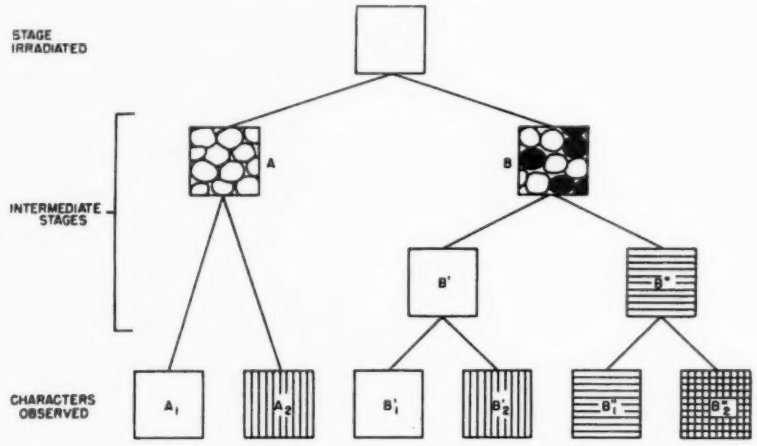

FIG. 2. Simplified diagrammatic representation of the possible pathways in the genesis of radiation-induced developmental abnormalities. It shows selectivity of early developmental damage as due to both (*a*) uneven distribution among precursor regions (A, B) of initial cellular damage (probably consisting of death of cells, see text) and (*b*) different degrees of regulatory powers of different processes (B′, B″). Thus, A_1 is normal because of lack of damage to precursors; while B′ and B_1' are normal in spite of damage to precursors. Following the initial localization (B″) of developmental damage, abnormalities may be produced in direct line of cellular descent or they may be secondarily caused. Thus, the genesis of B_1'' is directly and simply traceable to initial damage. Malformation A_2 is secondarily produced (e.g., by a condition set up by B″) in a character descended from undamaged tissue (A) The production of B_2' also is purely secondary, even though the cells ancestral to this character were damaged as a direct consequence of irradiation. Einally, abnormality B_2'' is a result of interaction between direct and secondary effects.

Open areas = no damage; ● = initial cellular damage; ▤ = developmental disturbance traceable by cellular descent to initial damage; ▥ = secondary damage; ▦ = both types of damage. [Courtesy of Russell & Russell (51).]

In another paper Russell & Russell (52) discuss pathways of radiation effects from two viewpoints: (*a*) genetic studies wherein the maternal germ cells are irradiated before fertilization and (*b*) developmental experiments in which the embryo is irradiated in various stages of development. The authors comment:

> We find ourselves at a rather inconvenient stage, at which the accumulation of data has become sufficiently extensive to enable us to speculate on many interesting and fundamental points, whereas the additional experiments designed to test these speculations have not yet been completed.

Some of the important considerations emerging from this study are as follows:

(*a*) The question of whether some of the responses of the embryo may be due to the concomitant irradiation of the mother has been answered largely in the negative, especially for the genetic experiments and for the induction of developmental abnormalities by irradiation of embryos.

(*b*) Applying certain findings from the general field of radiation genetics and histology, the hypothesis has been advanced that most if not all results may be explained by the radiation induction of nuclear damage (in maternal germ cells or in embryonic cells, as the case may be). Damage seems to be of two types: one, of unknown nature, expressing itself in nuclear breakdown at the first mitosis; the other being in the form of chromosome breaks which produce aberrations and lead to chromosome imbalance after cell division and, sooner or later, to death of descendent cells. There is some direct evidence for the more rapid type of cell death.

(*c*) The production of abnormalities, their dependence on developmental stage irradiated, and the differential sensitivity of parts of the embryo to developmental damage may be explained by cell death (possibly by both of the above mechanisms) differentially distributed and initiating developmental effects in the embryonic precursors in which it occurs if the regulatory powers of these precursors are insufficient to compensate for the given amount of cellular killing. Other abnormalities may later be produced secondarily by interaction effects.

It may be added that one of the especially important points emphasized in this paper, and supported by striking evidence, is that comparison of litter sizes does not measure radiation-induced mortality and the results of investigations in which this criterion has been used will have to be reappraised.

Finally Russell & Russell (53) in a paper on hazards to the embryo and fetus from ionizing radiation, emphasize the application of their results to the practical problems encountered when humans are irradiated. We believe it important to give here the author's summary to this paper:

> The results of some recent experiments on the effects of radiation on mammalian embryos are summarized and applied to the problem of radiation hazards to the human embryo. Experiments indicate that the potential radiation hazard is greatest during the period of major organogenesis. In a human pregnancy, at least part of this period is so early that pregnancy may still be unsuspected. It is, therefore, recommended that, whenever possible, pelvic irradiation of women of child-bearing age should be restricted to the two weeks following the menses, when there is little chance of an unsuspected pregnancy. This applies particularly to diagnostic fluoroscopy

which can be in the hazardous dose range. On the other hand, it seems unlikely that the present permissible weekly dose which may be received in occupational irradiation constitutes a measurable hazard at any stage in pregnancy.

As part of the ORNL Symposium Hicks (54) presented a paper on the effects of radiation, hormones, and radiomimetic drugs on the developing nervous system of rats and mice. It represented some highlights in the program which has been in progress in his laboratory for several years past. We think it important to set down the threefold purpose of his program: (*a*) to ascertain how various tissues, especially neural tissue, differ metabolically and, in turn, how they respond differently to different deleterious agents; (*b*) to determine how embryonic tissues differ, among themselves and from adult tissues, and how their metabolism changes during development; (*c*) to correlate specific "metabolic vulnerabilities" of normal tissues to problems of cancer chemotherapy and pathological embryology. As far as the limited content of this paper of his is concerned the following summary points are especially important:

Ionizing radiations at low doses selectively damage the primitive neuroblasts and spongioblasts in embryo, fetal, and young animals; and, experimentally, a series of reproducible patterns of malformations of the nervous system can be achieved based essentially on the destruction of these building blocks. The patterns range from anencephaly through hydrocephalus to various kinds of microcephaly and cerebellar defects. With the effective dose of radiation remaining constant, the pattern of malformation is determined by the time during development at which the radiation is given.

Here also Hicks has presented a timetable of radiation malformations with special emphasis on neural elements. He has indicated (by personal communication) that the timetable is something that "we keep improving." Accordingly we are pleased to present in Figure 3 a somewhat more up-to-date version courteously provided this reviewer by Dr. Hicks; in his letter he indicates that while the earlier (54) timetable is valid " . . . it does not emphasize the interesting 'recovery' of embryos in the 4 to 10 somite stage which results in defects virtually limited to the eyes."

Regarding radiomimetic drugs such as nitrogen mustard and triethylene melamine, and certain sulfhydryl reagents these

. . . selectively destroy the radiosensitive neuroblasts and spongioblasts . . . Varying degrees of success in producing malformations have accompanied their use, but the selectivity of action of the sulfhydryl reagents has been less than that of radiation . . .

In short,

The patterns of malformation that result from the action of various noxious agents on developing mammals are largely determined by the product of two factors: the character of the agent and the time of injury.

In a later study, Hicks (55) considers the following problem:

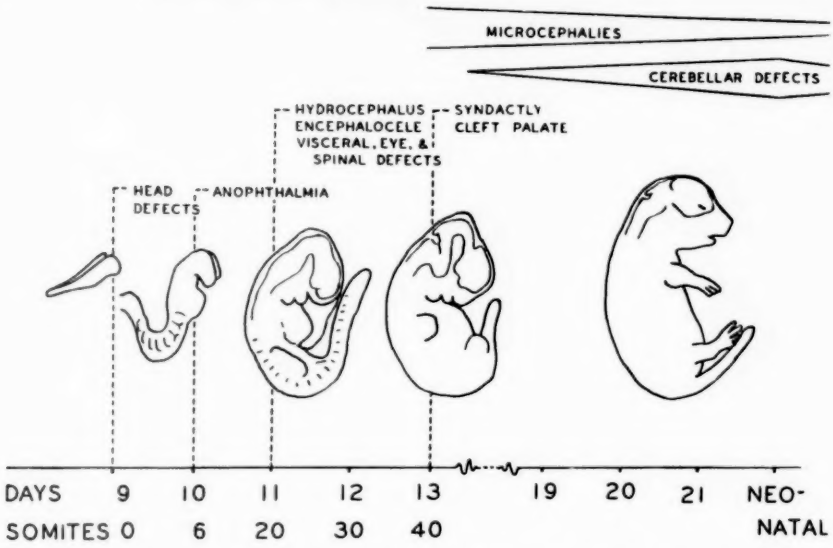

FIG. 3. A timetable of radiation malformation in rats and mice (slightly redrawn from an unpublished figure generously supplied for this review by Dr. Samuel P. Hicks). This figure embodies additional data not available at the time of the construction of the timetable presented in Hicks (57).

What has been especially puzzling in numerous past experiments has been how on the 9th day anencephaly could result, hydrocephalus, midbrain and skeletal deformities could occur on the 11th day; yet on the 10th day in between, anophthalmia was almost the only defect.

The striking character of this situation is illustrated in Figure 4. Experiments have revealed that at certain periods the mammalian embryo has a "tremendous capacity to recovery from injury." The report proposes to demonstrate this reparative process in the rat embryo, to show its importance, and to relate findings to past experiments. For the important improved experimental procedure accompanying this work the paper must be consulted. The substance of the principal outcome of this important investigation can be gathered from the following:

It had been previously established that malformations, especially of the nervous system, were closely associated with or initiated by the destruction of the nonmitotic differentiating primitive neuroblasts and spongioblasts which are selectively killed by low doses of radiation.

[See Hicks (54)]

This concept must now be modified by the present more precise experiments. These demonstrate that despite extensive necrosis of differentiating neural cells and cells in an analogous stage in other developing systems, complete or nearly complete repair can occur in many zones. Radiation malformations are therefore the result

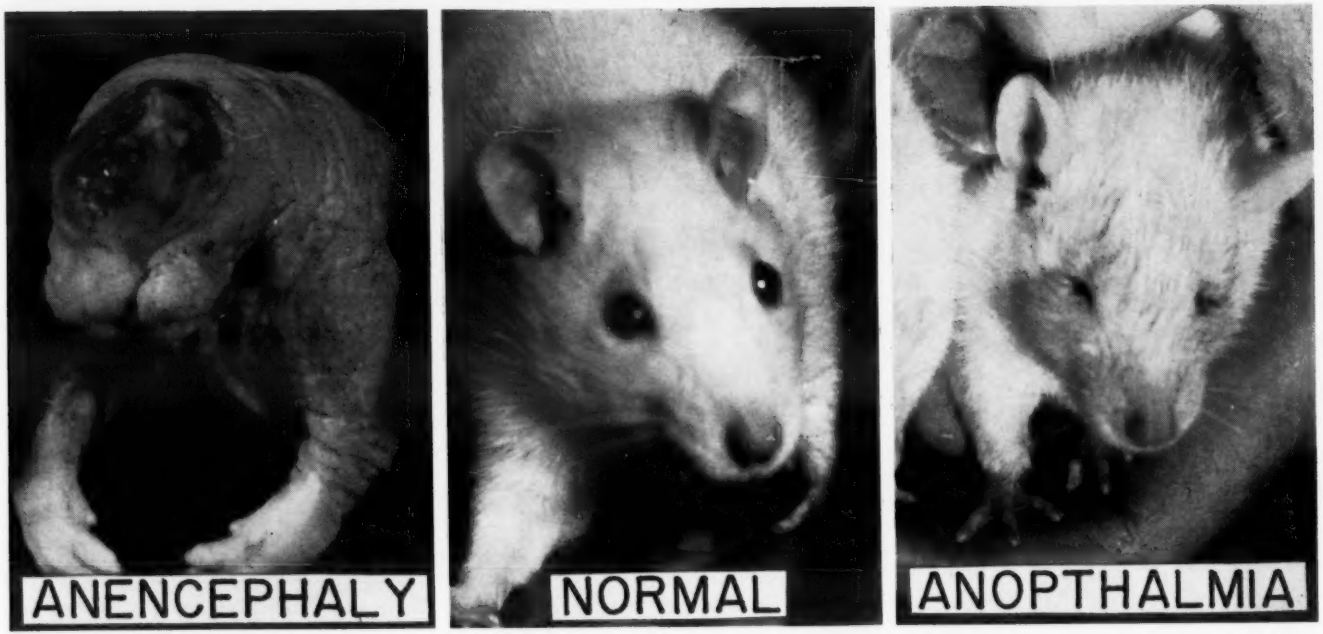

FIG. 4. Left, anencephaly in a newborn (Caesarian) rat resulting from irradiation on the ninth day of gestation. Right, anophthalmia in an adult rat resulting from irradiation on the tenth day of gestation; but for the anophthalmic condition this animal was virtually normal. [Courtesy Hicks (58).]

of a balance between radiation damage and the capacity to repair in any given anlage or developing zone.

In the recent *Biochemistry of the Developing Nervous System*, Hicks (56) has contributed a brief section on the neurochemical significance of experimentally-induced malformations in mammals; the material here is contained in his previous publications.

In addition, although this is not primarily a radiodevelopmental study, Hicks and his associates (57) have recently engaged the further problem of time-intensity factors in radiobiological response and here too the emphasis is on the central nervous system. Their initial study in this category concerns acute effects of megavolt electrons and high and low energy x-rays (215 Mev electrons and 150 Kv-3.0 Mev x-rays) on the brain of mice (principally) and rats. A few pregnant animals were used to compare these effects with effects of other radiations on the fetus. [In a preliminary report, using these types of radiations, Hicks & Wright (58) found that very high intensity megavolt electrons not only could destroy grey and white matter of the brain but could kill the animal instantly when the dose was sufficiently high.] They write: "Altogether, the intensity of the radiation—the time in which the dose was given—emerged as an important factor in determining severity of brain damage."

In the ORNL Symposium (3) there appear two papers by distinguished embryologists, B. H. Willier and P. Weiss. Willier's (64) contribution on phases in the time scale of the development of the chick embryo was especially timely considering the extent to which the concept of "critical periods" was discussed; he writes:

> Each major phase in development apparently represents a transition from one state to another, irrespective of whether the change is structural or functional in nature. Furthermore, each major phase seemingly comprises a series of less conspicuous steps, any one of which might be regarded as a critical or sensitive phase for the moment of action of the deleterious agents.

Weiss (65), chairman of the program, made important summarizing remarks; among other things he points out that if certain fundamental embryological considerations are kept in mind there will be "no danger of misinterpreting such terms as 'critical phase' or 'sensitive periods' as if they were sharply demarcated phases."

Clinical

It may be stated that, in principle, any and all observations on the effects of ionizing radiations on living systems have at least some implications (recognized or not) for medical practice. The importance, in this regard, of studies of the kind described above, and particularly those on mammals, hardly needs emphasizing. Before entering upon a brief review of some recent studies on humans exposed *in utero* to atomic bomb explosions we

should like to call attention again to the paper [discussed in Rugh (1)] by Russell & Russell (59) on radiation hazards to the embryo and fetus which is directed particularly to implications (from experimental studies) for medical practice. In this connection (although we have excluded genetic considerations from this review) we should like to recommend a small paper by Rugh (60) on genetic hazards in ovarian irradiation.

Plummer (61) investigated anomalies occurring in children exposed while *in utero* to the atomic explosion in Hiroshima. The study embraced 205 children, then four and one-half years old, who had been exposed to the atomic blast during the first half of their intra-uterine life; eleven of these were within 1200 meters of the bomb hypocenter[8] and the rest somewhat beyond that distance. Seven of the eleven children exposed within 1200 meters exhibited microcephaly and mental retardation; this diagnosis was not made on any of the other 194 children who were at greater distances from the hypocenter. The author concludes from this study that defects in the central nervous system can be produced in a human fetus by atomic bomb radiation, if exposure occurs within approximately 1200 meters of the hypocenter and the fetus has not been effectively shielded at the same time (as by concrete) from direct exposure to radiation.

There have also been studies on children exposed *in utero* when the atom bomb was dropped on Nagasaki. Sutow & West (62) made a roentgenographic survey of the occurrence of skeletal malformations of 74 children exposed *in utero* at distances less than 2000 meters from hypocenter; at the time of this study these children were between six and seven years of age; the observations on this group were compared with another group of 91 children ("controls") who were also exposed *in utero* to the same bomb but at distances of 4000 to 5000 meters from hypocenter. Approximate gestation ages at exposure were calculated from birth dates and classified in terms of "probable trimester of pregnancy." For a proper appreciation of the extent of this postirradiation roentgenological appraisal of skeletal malformations the original paper must be consulted. In short, no difference was found in the incidence of more common bone variations in the above two groups of children. Skeletal abnormalities such as those found in experimentally irradiated rats and mice were not found. The authors comment: "It is quite possible that the more extreme degrees of abnormalities were present among the fetuses that were aborted or delivered dead and among infants that died in the early postnatal period." In this study an effort was also made to ascertain whether the presence of radiation symptoms in the mother was associated with higher incidence of skeletal abnormalities in the offspring; the numbers of individuals involved in this study were small and no correlation of these two factors could be established.

Yamazaki, Wright & Wright (63) addressed themselves to the problem

[8] Hypocenter is generally defined as that point on the ground directly over which the bomb is calculated to have exploded.

of the outcome of pregnancies in women exposed to the atomic blast at Nagasaki; their observations were presented at the ORNL Symposium to which reference has been made. They found that among 30 mothers with one or more major manifestations of radiation damage (epilation, oropharyngeal lesions, purpura, petechiae), who were within 2000 meters of hypocenter, there were seven fetal deaths (23.4 per cent), six neonatal and infant deaths (26 per cent), four cases of mental retardation among 16 surviving children (25 per cent). There was thus an overall morbidity and mortality in approximately 60 per cent of this group of mothers. This stands in remarkable contrast to a group of 68 mothers who were also within 2000 meters of hypocenter but were without manifestations of major radiation damage; in this group the overall mortality was only 10 per cent. In the "control" group (4000 to 5000 meters from hypocenter) the overall mortality was only about six per cent. It is also important to note that the mean height and head circumference of children born to mothers who exhibited major signs of radiation damage were significantly smaller than in children born to mothers belonging to the "control" group. The authors submit that, in spite of difficulties attending evaluation of data the evidence strongly suggests that radiation either directly to the fetus, or indirectly as a result of its effect on maternal tissues, was largely important "in determining the results of these pregnancies."

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